

Literature Cited

- (1) Perry, J. H. "Chemical Engineer's Handbook", 3rd ed.; McGraw-Hill: New York, 1950; p 166.
- (2) Zelsberg, F. C. *Chem. Metall. Eng.* **1925**, *32*, 326.
- (3) Fritz, J. J.; Fuget, C. R. *Ind. Eng. Chem., Chem. Eng. Ser.* **1959**, *1*, 10.
- (4) Othmer, D. F.; Naphtali, L. M. *Ind. Eng. Chem., Chem. Eng. Ser.* **1959**, *7*, 1.
- (5) Harned, H. S.; Ehlers, R. W. *J. Am. Chem. Soc.* **1932**, *54*, 1350.
- (6) Harned, H. S.; Ehlers, R. W. *J. Am. Chem. Soc.* **1933**, *55*, 2179.
- (7) Akerlof, G.; Teare, J. W. *J. Am. Chem. Soc.* **1937**, *59*, 1855.
- (8) Rupert, F. F. *J. Am. Chem. Soc.* **1909**, *31*, 851.
- (9) Haase, R.; Naas, H.; Thumm, H. Z. *Phys. Chem. (Frankfurt am Main)* **1963**, *37*, 210.
- (10) Kao, J. T. F. *J. Chem. Eng. Data* **1970**, *15*, 362.
- (11) Dunn, J. S.; Rideal, E. K. *J. Chem. Soc.* **1924**, 125, 676.
- (12) Van Ness, H. C. *AIChE J.* **1970**, *16*, 18.

Received for review May 24, 1982. Revised manuscript received April 11, 1983. Accepted April 29, 1983. This work was supported by the U.S. Air Force Office of Scientific Research under Grant AFOSR-77-3333.

Measurements of Densities and Dielectric Constants of Liquid Isobutane from 120 to 300 K at Pressures to 35 MPa

William M. Hayn

Thermophysical Properties Division, National Engineering Laboratory, National Bureau of Standards, Boulder, Colorado 80303

Measurements of the densities and dielectric constants of compressed liquid isobutane have been carried out at temperatures between 120 and 300 K to pressures of 35 MPa. These experimental data along with computed values for the Clausius-Mossotti function (CM) are reported in this paper.

Introduction

This work is part of a large-scale program at this laboratory to determine the thermophysical properties of technically important fluids. In addition to isobutane being a major component of petroleum and natural gases, it is a prime candidate as a working fluid in geothermal energy processes. This work was undertaken to provide density and dielectric constant data for isobutane in regions not previously investigated.

Experimental Section

Detailed descriptions of the experimental apparatus and procedures have been presented in other papers (1-5). Only information essential to understand this paper is presented here.

Simultaneous measurements of density and dielectric constant were made on the same liquid samples. A magnetic suspension densimeter was used to obtain the density data, while a concentric cylindrical capacitor contained in the same apparatus was employed for the dielectric constant measurements. Pressures were measured with an oil-operated dead-weight gauge. The isobutane sample was separated from the oil by a diaphragm-type differential pressure indicator. The primary temperature sensor was a platinum resistance thermometer.

For the magnetic suspension densimeter used in the present work, the magnetic moment of the float material is a relatively strong function of temperature. Thus, it is most practical to take data along isotherms. For each temperature, the cell was first filled to the highest pressure (35 MPa) for that run. Data points at lower pressures were taken after venting appropriate amounts of gas. For each isotherm the last point was taken on the coexistence boundary to compare with previous saturated-liquid density results (6) obtained with a magnetic suspension densimeter in this laboratory. The present saturated-liquid data agreed with those taken earlier to better than 0.02% and are not published here since they were used only as a

check on the present work. (Saturated-liquid dielectric constants obtained in the present study have been published elsewhere (5).) Vacuum measurements, needed at each temperature for absolute density measurements with the magnetic suspension densimeter used here, were obtained, for each run, before charging the cell with liquid isobutane.

The samples were obtained from cylinders of research-grade, commercially available liquid isobutane. The minimum purity as specified by the supplier was 99.90 mol %, with the most probable impurity being *n*-butane.

Results and Discussion

The experimental densities (ρ) and dielectric constants (ϵ) for liquid isobutane are given as a function of temperature (T , IPTS-68) and pressure (P) in Table I. Also presented in this table are values for the Clausius-Mossotti function (CM) calculated from the relation

$$\text{CM} = \left(\frac{\epsilon - 1}{\epsilon + 2} \right) \frac{1}{\rho} \quad (1)$$

Data have been obtained for 11 isotherms between 120 and 300 K. Two of the isotherms (at 140 and 160 K) were repeated on new samples to determine the reproducibility of the measurements. Each isotherm was comprised of 12 data points taken at pressures approximately equal to those for any other isotherm. The density range for this work extended from 551 kg/m³, or approximately 2.5 times the critical density, to 749 kg/m³, which is slightly greater than the triple-point density.

The estimated total uncertainty in the experimental densities is less than 0.1% while the imprecision of the measurements is a few parts in 10⁴ (3). The estimate of the imprecision was substantiated by the reproducibility of the isotherms at 140 and 160 K on different liquid samples. Capacitance measurements to a resolution of 10⁻⁴ pF, combined with better than 10⁻⁴-pF stability in the vacuum capacitance, gave an estimated uncertainty of approximately 0.01% in the dielectric constant. For pressure, the overall uncertainty was approximately 0.01%, increasing somewhat at lower pressures. Temperatures were measured to a precision of a few mK; however, the total uncertainty could be as large as 30 mK at 300 K, decreasing to approximately 15 mK at 120 K.

The density and dielectric constant data from this work have been used in comprehensive correlations (7, 8) of the thermophysical properties of isobutane. Only one other set of data

Table I. Experimental Densities (ρ), Dielectric Constants (ϵ), and Clausius-Mossotti Functions (CM) of Liquid Isobutane as a Function of Pressure (P) and Temperature (T , IPTS-68)

P/MPa	$\rho/(\text{kg m}^{-3})$	ϵ	CM/ ($\text{cm}^3 \text{mol}^{-1}$)	P/MPa	$\rho/(\text{kg m}^{-3})$	ϵ	CM/ ($\text{cm}^3 \text{mol}^{-1}$)
$T = 120.000 \text{ K}$							
34.7208	748.89	2.116 92	21.0567	14.0618	740.95	2.104 00	21.1024
31.2775	747.60	2.114 84	21.0643	10.6189	739.61	2.101 73	21.1089
27.8345	746.29	2.112 73	21.0722	7.8644	738.51	2.099 88	21.1142
24.3914	744.97	2.110 59	21.0798	5.1102	737.36	2.098 02	21.1209
20.9480	743.63	2.108 42	21.0878	3.0443	736.52	2.096 61	21.1252
17.5050	742.29	2.106 22	21.0952	1.6672	735.97	2.095 66	21.1276
$T = 130.000 \text{ K}$							
34.7197	740.21	2.096 97	21.0248	14.0603	731.74	2.083 16	21.0717
31.2765	738.81	2.094 74	21.0336	10.6174	730.30	2.080 75	21.0785
27.8330	737.39	2.092 49	21.0419	7.8629	729.14	2.078 77	21.0836
24.3898	735.99	2.090 21	21.0500	5.1086	727.98	2.076 77	21.0885
20.9465	734.58	2.087 90	21.0575	3.0428	727.08	2.075 26	21.0925
17.5035	733.17	2.085 55	21.0645	1.6657	726.47	2.074 24	21.0959
$T = 140.000 \text{ K}$							
34.7206	731.59	2.077 81	20.9995	34.7188	731.35	2.077 82	21.0064
31.2773	730.60	2.075 47	21.0095	31.2755	730.02	2.075 46	21.0107
27.8337	728.60	2.073 08	21.0174	27.8319	728.55	2.073 07	21.0186
24.3905	727.13	2.070 65	21.0248	24.3887	727.08	2.070 65	21.0260
20.9471	725.66	2.068 18	21.0315	20.9453	725.59	2.068 17	21.0333
17.5039	724.35	2.065 68	21.0331	17.5022	724.10	2.065 66	21.0402
14.0607	722.78	2.063 13	21.0417	14.0590	722.58	2.063 11	21.0472
10.6179	721.15	2.060 54	21.0511	10.6161	721.00	2.060 52	21.0550
7.8631	719.87	2.058 41	21.0573	7.8616	719.75	2.058 40	21.0606
5.1089	718.59	2.056 27	21.0632	5.1073	718.50	2.056 26	21.0658
3.0429	717.61	2.054 64	21.0680	3.0416	717.56	2.054 63	21.0690
1.6658	716.97	2.053 54	21.0706	1.6645	716.92	2.053 52	21.0715
$T = 160.000 \text{ K}$							
34.7211	714.54	2.041 42	20.9616	34.7198	714.82	2.041 43	20.9534
31.2776	712.91	2.038 78	20.9699	31.2765	713.16	2.038 77	20.9626
27.8341	711.23	2.036 08	20.9788	27.8330	711.54	2.036 07	20.9696
24.3910	709.55	2.033 34	20.9871	24.3899	709.82	2.033 33	20.9790
20.9476	707.81	2.030 53	20.9960	20.9465	708.05	2.030 52	20.9890
17.5045	706.09	2.027 67	21.0039	17.5034	706.27	2.027 67	20.9982
14.0613	704.30	2.024 75	21.0125	14.0602	704.50	2.024 75	21.0064
10.6184	702.50	2.021 78	21.0210	11.3059	703.05	2.022 37	21.0135
7.8639	701.08	2.019 35	21.0261	8.5513	701.54	2.019 95	21.0214
5.1097	699.65	2.016 88	21.0307	5.7971	700.09	2.017 49	21.0270
3.0438	698.54	2.014 99	21.0348	3.7314	698.93	2.015 60	21.0328
1.6667	697.80	2.013 72	21.0377	1.6655	697.82	2.013 71	21.0369
$T = 180.000 \text{ K}$							
34.7205	698.14	2.006 91	20.9217	14.0609	686.35	1.987 97	20.9800
31.2771	696.32	2.003 92	20.9297	11.3066	684.66	1.985 23	20.9877
27.8336	694.45	2.000 87	20.9382	8.5521	682.93	1.982 44	20.9963
24.3905	692.53	1.997 76	20.9472	5.7978	681.19	1.979 59	21.0038
20.9472	690.51	1.994 57	20.9580	3.7322	679.89	1.977 40	21.0084
17.5041	688.44	1.991 32	20.9694	1.6663	678.53	1.975 19	21.0144
$T = 200.000 \text{ K}$							
34.7199	681.48	1.974 07	20.9053	14.0603	668.06	1.952 55	20.9679
31.2766	679.39	1.970 72	20.9154	11.3061	666.10	1.949 40	20.9764
27.8331	677.29	1.967 28	20.9240	8.5516	664.13	1.946 16	20.9841
24.3899	675.08	1.963 74	20.9340	5.7974	662.14	1.942 85	20.9914
20.9466	672.84	1.960 12	20.9441	3.7317	660.58	1.940 30	20.9975
17.5036	670.50	1.956 39	20.9554	1.6659	658.98	1.937 70	21.0042
$T = 220.000 \text{ K}$							
34.7200	665.12	1.942 23	20.8869	14.0605	649.64	1.917 77	20.9593
31.2767	662.62	1.938 45	20.9017	11.3062	647.36	1.914 11	20.9690
27.8332	660.21	1.934 55	20.9114	8.5517	645.03	1.910 37	20.9786
24.3901	657.76	1.930 55	20.9209	5.7974	642.61	1.906 50	20.9889
20.9467	655.16	1.926 26	20.9298	3.7318	640.78	1.903 51	20.9956
17.5037	652.45	1.922 17	20.9457	1.6659	638.95	1.900 44	21.0006
$T = 240.000 \text{ K}$							
34.7206	648.35	1.911 36	20.8885	14.0611	630.96	1.883 54	20.9582
31.2773	645.65	1.907 12	20.9011	11.3068	628.31	1.879 33	20.9691
27.8339	642.90	1.902 73	20.9126	8.5529	625.59	1.874 95	20.9792
24.3908	640.10	1.898 18	20.9224	5.7980	622.80	1.870 40	20.9883
20.9474	637.19	1.893 50	20.9335	3.7323	620.67	1.866 86	20.9938
17.5043	634.17	1.888 66	20.9454	1.6665	618.47	1.863 21	20.9993

Table I (Continued)

P/MPa	$\rho/(\text{kg m}^{-3})$	ϵ	CM/ ($\text{cm}^3 \text{mol}^{-1}$)	P/MPa	$\rho/(\text{kg m}^{-3})$	ϵ	CM/ ($\text{cm}^3 \text{mol}^{-1}$)
$T = 260.000 \text{ K}$							
34.7194	631.93	1.881 19	20.8831	14.0602	612.16	1.849 53	20.9540
31.2761	628.90	1.876 44	20.8960	11.3060	609.14	1.844 61	20.9625
27.8327	625.80	1.871 50	20.9081	8.5515	605.97	1.839 47	20.9719
24.3897	622.58	1.866 38	20.9203	5.7973	602.60	1.834 09	20.9834
20.9464	619.25	1.861 03	20.9318	3.7317	599.96	1.829 87	20.9921
17.5033	615.77	1.855 41	20.9432	1.6658	597.22	1.825 44	21.0006
$T = 280.000 \text{ K}$							
34.7209	614.96	1.851 62	20.8985	14.0611	592.30	1.815 42	20.9725
31.2775	611.59	1.846 27	20.9106	11.3069	588.73	1.809 65	20.9822
27.8340	608.03	1.840 69	20.9246	8.5523	585.03	1.803 55	20.9895
24.3908	604.35	1.834 85	20.9379	5.7981	581.14	1.797 09	20.9957
20.9474	600.52	1.828 73	20.9503	3.7324	578.03	1.791 96	21.0012
17.5043	596.52	1.822 26	20.9616	1.6665	574.76	1.786 55	21.0064
$T = 300.000 \text{ K}$							
34.7216	598.61	1.822 67	20.8965	14.0620	572.38	1.781 19	20.9796
31.2783	594.84	1.816 67	20.9081	11.3077	568.15	1.774 33	20.9886
27.8348	590.88	1.810 36	20.9204	8.5531	563.71	1.767 01	20.9943
24.3916	586.64	1.803 70	20.9352	5.7989	558.89	1.759 11	21.0015
20.9483	582.16	1.796 68	20.9506	3.7332	554.91	1.752 75	21.0104
17.5052	577.41	1.789 18	20.9655	1.6674	550.70	1.745 95	21.0178

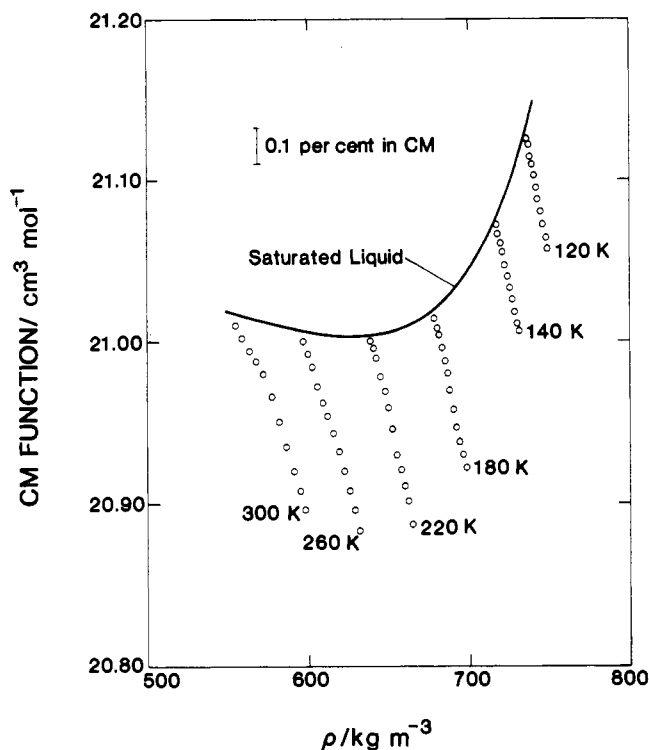


Figure 1. Clausius-Mossotti function as a function of density for liquid isobutane; data from this work and ref 5.

overlap the results of the present work. Sage and Lacey (9) obtained data for isobutane at temperatures between 294 and 394 K at pressures to 20 MPa. When compared via the equation of state in ref 7, the densities of Sage and Lacey at 294 K were approximately 1% larger than those from this work.

In Figure 1 the Clausius-Mossotti function is plotted as a function of density for selected isotherms for isobutane. The saturated-liquid values were taken from an earlier paper (5). At low temperatures (or high densities) along the saturation curve, the CM function increased with decreasing temperature. This behavior is the opposite of that exhibited by *n*-butane (5), for which the CM function decreases with increasing density (decreasing temperature) for a density range from approximately the critical density to the triple-point liquid density. The behavior exhibited by *n*-butane is typical of that for most simple, nonpolar molecules. Isobutane and also propane (5) are very weakly polar fluids with small dipole moments (μ) and are characterized by the sharp increase, proportional to μ^2/T , in the CM function at low temperatures for the liquid along the saturation curve.

Registry No. Isobutane, 75-28-5.

Literature Cited

- (1) Haynes, W. M. *J. Res. Natl. Bur. Stand. (U.S.)*, in press.
- (2) Haynes, W. M. *J. Chem. Thermodyn.* 1982, 14, 803.
- (3) Haynes, W. M.; Hiza, M. J.; Frederick, N. V. *Rev. Sci. Instrum.* 1976, 47, 1237.
- (4) Haynes, W. M. *Rev. Sci. Instrum.* 1977, 48, 38.
- (5) Haynes, W. M.; Younglove, B. A. In "Advances in Cryogenic Engineering"; Fast, R. W., Ed.; Plenum Press: New York, 1982; Vol. 27, p 883.
- (6) Haynes, W. M.; Hiza, M. J. *J. Chem. Thermodyn.* 1977, 9, 179.
- (7) Goodwin, R. D.; Haynes, W. M. *NBS Tech. Note (U.S.)* 1982, No. 1051.
- (8) Waxman, M.; Gallagher, J. *J. Chem. Eng. Data*, in press.
- (9) Sage, B. H.; Lacey, W. N. *Ind. Eng. Chem.* 1938, 30, 673.

Received for review November 10, 1982. Accepted March 11, 1983. This work was carried out at the National Bureau of Standards under the sponsorship of the Gas Research Institute.