

Viscosity of Hydrocarbons. Propane

L. T. CARMICHAEL, VIRGINIA M. BERRY, and B. H. SAGE
California Institute of Technology, Pasadena, Calif.

Investigations of the viscosity of propane were carried out in the liquid and gas phases at pressures up to 5000 p.s.i.a. in the temperature interval between 40° and 400° F. These measurements serve to corroborate and extend the results reported in more extensive investigations carried out with entirely different types of instruments. For the most part, satisfactory agreement between several sets of investigations has been found. However, earlier measurements carried out with a rolling-ball viscometer indicate that the latter type of instrument is entirely unsuitable for measurements in the gas phase. Utilizing the current experimental data obtained for propane along with those which have recently been recorded in connection with the viscosity of ethane and *n*-butane, a comparison was made with the data reported by several investigators using various types of instruments.

THE VISCOSITY OF PROPANE has been investigated in some detail. Trautz and Sorg (20) carried out a limited investigation of propane at atmospheric pressure at temperatures from 64° to 483° F., while Smith and Brown (12) studied the viscosity of propane at pressures up to 5000 p.s.i.a. at temperatures between 71° and 342° F. Comings and coworkers (5) studied the viscosity of propane at pressures up to approximately 600 p.s.i.a. at temperatures between 86° and 220° F. and devoted some effort to preparing a correlation of the viscosity of gases at elevated pressures. Baron and coworkers (1) made a rather complete investigation of the viscosity of propane at pressures up to 8000 p.s.i.a. in the temperature interval between 125° and 275° F., while Bicher and Katz (2) measured the viscosity of mixtures of methane and propane as well as the viscosity of the pure components. This latter work was carried out at pressures up to 5000 p.s.i.a. at temperatures between 77° and 437° F. Starling, Eakin, and Ellington (15) recently carried out an additional investigation of the viscosity of propane at pressures up to 8000 p.s.i.a. in the temperature interval between 70° and 460° F. Earlier Sage and Lacey (10) investigated the viscosity of propane with a rolling-ball instrument at pressures up to 2000 p.s.i.a. in the temperature interval between 100° and 220° F. The data for the gas phase at elevated pressures were found to be unsatisfactory as a result of the relatively high Reynolds numbers experienced in the viscometer. However, the measurements in the liquid phase appear to be in reasonable agreement with more recent data.

Experience has indicated that before confidence can be placed in viscosity measurements, it is desirable to obtain measurements over a range of temperatures and pressures from at least two widely different types of instruments. With this in view, the viscosity of propane measurements were undertaken to strengthen confidence in the values reported by Starling and coworkers (15). The measurements of Baron, Roof, and Wells (1) and Starling and coworkers (15) were made in similar types of instruments and thus do not afford the desirable comparison of data obtained with widely different types of instruments. For this reason, a limited number of measurements were carried out at pressures up to 5000 p.s.i.a. in the temperature interval between 40° and 400° F.

METHODS AND EQUIPMENT

The equipment employed for this investigation was of the rotating-cylinder type, and the details of the apparatus have been presented (9) with the modifications found desirable (3). The methods utilized were the same as

those employed in an earlier study of ethane (3). In principle, the apparatus consisted of a rotating cylinder within which a stator was suspended on a small platinum-tungsten wire. Measurements of the angular displacement of the stator as a result of the rotation of the outer cylinder at a predetermined angular rate permitted the viscosity of the fluid in the gap to be evaluated from the dimensions of the instrument and the elastic characteristics of the suspension.

The elastic characteristics of the suspension were determined as a function of time and temperature from the natural period of the stator. The angular displacement of the stator was determined by means of an angular transducer of the reluctance type (3). Measurements were carried out with the cylinder rotating in each direction. The speed of the outside cylinder was established by a predetermined drive and was known within 0.01% while the elastic characteristics were known at a prescribed temperature within 0.1%. The dimensions of the stator and the rotating cylinder were checked periodically and were sufficiently stable at this stage in the life of the apparatus that the gap between the stator and the rotating cylinder did not change by more than 0.1% within the period covered by this investigation. The temperature was known within 0.05° F. relative to the international platinum scale. The uncertainty in pressure was 0.05% or 0.1 p.s.i., whichever is the larger measure of uncertainty.

MATERIAL

The propane used in this investigation was obtained as research grade from the Phillips Petroleum Co. The original material was reported to contain not more than 0.001 mole fraction of impurities. A mass spectrographic analysis indicated as much as 0.0028 mole fraction of compounds other than propane. The sample was purified by partial condensation techniques and re-evaporated at the temperature of solid carbon dioxide. The purified material yielded a change in vapor pressure of 0.29 p.s.i. with a change in quality from 0.1 to 0.9 at 160° F. From these measurements it is estimated that the sample of propane employed did not contain more than 0.0015 mole fraction of impurities which were for the most part other hydrocarbons containing three carbon atoms per molecule.

RESULTS

The experimental results, based solely upon the dimensions of the instrument and the elastic characteristics of the suspension, are set forth in Table I. Each measure-

ment was repeated at least two times with the rotor turning in both the forward and reverse directions. The results were averaged, and in no case did the individual measurements deviate from the average by more than 0.1%. The effect of pressure upon the viscosity of propane in the gas phase as established from the current measurements is shown in Figure 1. The corresponding measurements for the liquid phase constitute Figure 2. It should be remembered that these measurements were carried out with an entirely different type of instrument with the primary objective of checking the accuracy of earlier data. The standard error of estimate of the measurements for the liquid and gas phases was 1.53 micropoises. The residual viscosity of propane is shown in Figure 3. The standard error of estimate of the measured values from the smooth curve at specific weights greater than 10 pounds per cubic foot was 5.04 micropoises. This value assumes no uncertainty in the specific weight, and all the error lay in the measured viscosity.

COMPARISON WITH OTHER INVESTIGATORS

The viscosity of propane at atmospheric pressure as a function of temperature is shown in Figure 4. The standard error of estimate of each of the sets of data of other investigators (2, 5, 10, 15, 18, 19, 20) from the smooth curve has been indicated. For the most part the agreement is satisfactory. A comparison of the present measurements in the gas phase with the recommended values recorded by Starling and coworkers (15) is presented in Figure 5, along with the experimental data reported by Starling in his thesis (13) and by Comings and coworkers (5). Earlier

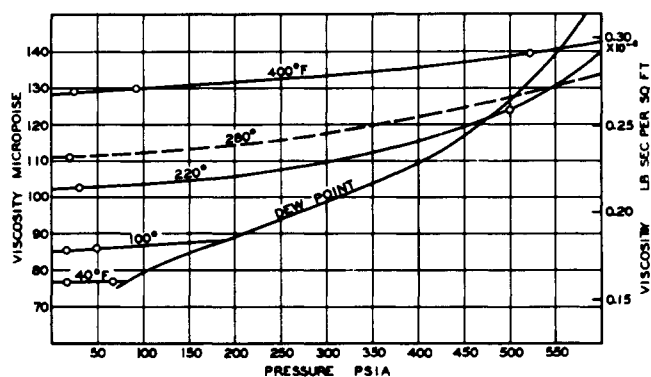


Figure 1. Viscosity of propane in the gas phase

measurements carried out with a rolling-ball viscometer (10) have not been included since the rapid increase in Reynolds number with pressure yielded a turbulent wake and variable points of separation around the ball, thus rendering measurements in the gas phase at elevated pressures unreliable. A comparison of available information for the dew-point gas (5, 13-15) is also presented in Figure 5.

Figure 6 shows the authors' data in the liquid phase as compared with other reported values (5, 13, 15). The data from the rolling-ball viscometer (10) have been included and yield fair agreement with the measurements of Starling (13). Utilizing several sets of data (8, 10, 13-17), a comparison for the bubble-point liquid is also shown in Figure 6.

The data for dew point and bubble point, for the most part, represent extrapolations to vapor pressure from measurements made in the single-phase regions. The present values and the recommended values of Starling and coworkers (15) are in reasonable agreement but greater disagreement is found between the data reported by Starling (13) and the authors' values.

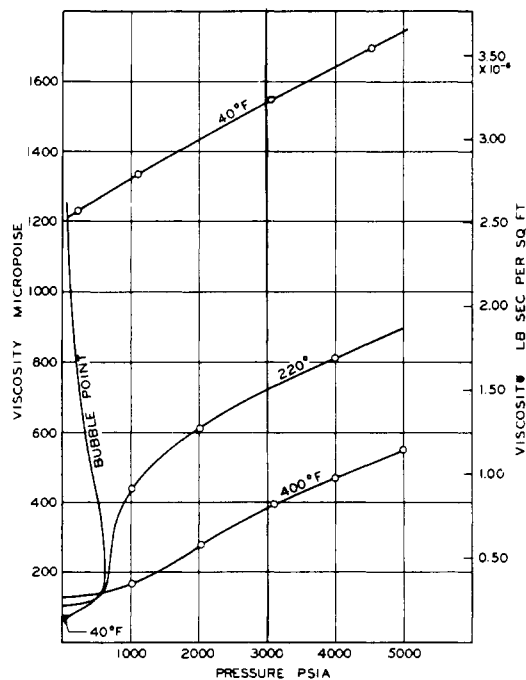


Figure 2. Viscosity of propane in the liquid phase

Table I. Experimental Measurements for Propane

Press., P.S.I.A.	Viscosity Micropoise	Press., P.S.I.A.	Viscosity Micropoise	Press., P.S.I.A.	Viscosity Micropoise
40° F.		220° F.		400° F.	
16.2	76.46	29.7	102.51	24.0	129.08
16.2	76.51	29.7	102.65	24.0	129.18
66.3	76.41	499.1	123.24	91.9	129.70
66.3	76.41	499.1	124.77	91.9	130.23
223.3	1229.82	1016.0	439.86	521.0	139.62
226.8	1233.84	1016.8	438.09	521.4	139.66
1113.3	1337.45	2010.4	611.34	1019.2	166.62
1117.6	1335.72	2011.9	611.08	1019.8	166.48
3053.6	1547.78	4001.9	810.97	2029.7	277.89
3065.6	1546.96	4010.5	811.69	2030.0	277.55
4512.8	1697.07			3093.2	392.61
4526.6	1696.06			3101.3	393.90
				3107.3	394.24
100° F.		280° F.			
16.0	85.22	19.9	110.94	4007.1	463.42
16.0	85.27	19.9	111.13	3974.9	471.22
16.0	85.32	19.9	111.27	3985.0	471.85
48.9	85.94			4969.8	548.50
48.9	86.08			4981.1	552.76
48.9	86.99			5028.7	548.69

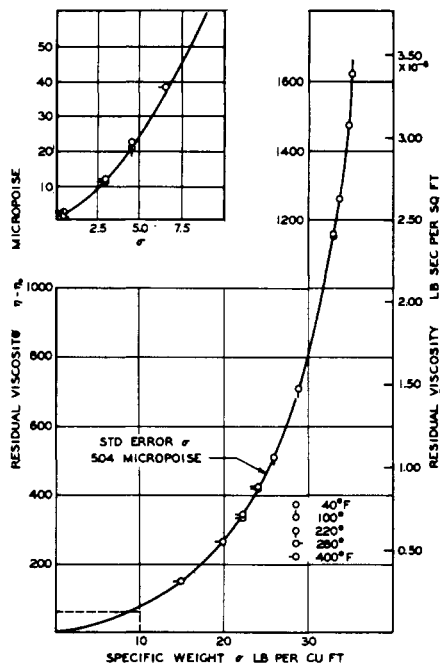


Figure 3. Residual viscosity of propane

In Table II are set forth smooth values of the viscosity of propane based in part upon the information submitted by Starling and coworkers (15) as well as upon earlier data modified and extended in range to reflect the trends found in the current measurements. The maximum disagreement between the information reported (15) and the current critically chosen values was 9.58 micropoises in the liquid phase and 2.73 micropoises in the gas phase both above and below the critical temperature of propane.

Sufficient experimental information has been accumulated concerning the viscosity of ethane, propane, and *n*-butane to warrant several simple comparisons. Utilizing a few conventional least squares techniques, the effect of temperature upon the viscosity at attenuation of these three paraffin hydrocarbons is shown in Figure 7. Corresponding values of the average deviation and standard deviation of the experimental points are set forth in Table III. The least squares techniques were applied, assuming that the

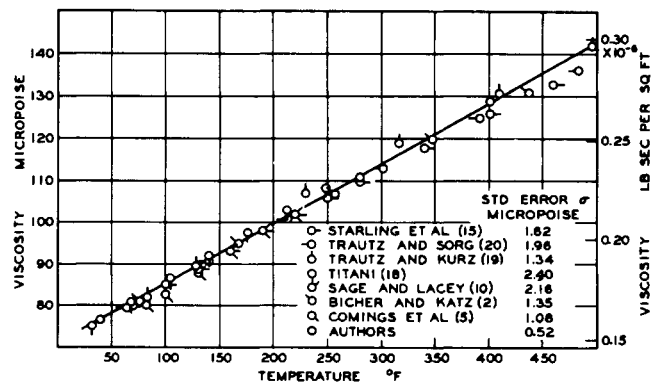


Figure 4. Viscosity of propane at atmospheric pressure

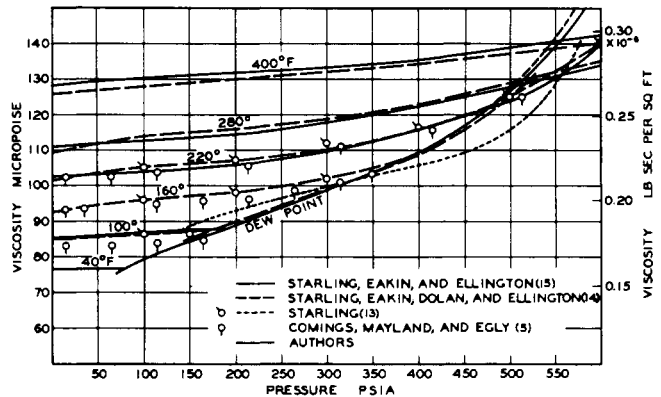


Figure 5. Data from several investigators for propane in the gas phase

effect of temperature upon the viscosity of these hydrocarbons at attenuation could be described by a simple polynomial of the following form:

$$\eta_0 = A + BT + CT^2 + DT^3 \quad (1)$$

The viscosity at attenuation was obtained by direct graphical extrapolation from values measured at atmospheric and higher pressures. The values of the coefficients of Equation 1 are set forth in a portion of Table III. Since, in the case of ethane, much closer agreement at attenuation can be obtained by elimination of Starling's values, this has been done in the second line of the tabulation which

Table II. Viscosity of Propane

Pressure P.S.I.A.	Temperature, ° F.						
	40	100	160	220	280	340	400
	(79) ^a	(188.7)	(383.8)				
Dew Point	76.51	87.86	107.63				
Bubble Point	1211.82	844.58	558.00				
14.7	76.51 ^b	85.13	93.94	102.56	111.13	119.70	128.41
200	1228.57	847.94	97.05	105.72	114.33	122.95	131.57
400	1253.47	877.14	562.00	115.96	122.33	128.79	135.49
600	1277.41	904.91	610.00	140.00	133.82	135.59	142.49
800	1300.87	932.20	650.00	345.02	157.00	148.00	152.26
1000	1323.85	956.14	682.00	433.02	207.99	169.00	165.18
1500	1378.91	1016.95	753.00	541.03	362.01	253.00	210.67
2000	1434.45	1072.49	815.00	597.05	452.98	335.00	273.87
2500	1489.04	1124.68	870.00	670.31	521.45	402.00	331.80
3000	1541.22	1175.91	922.00	722.97	574.98	460.00	383.99
3500	1593.41	1225.70	969.07	766.54	622.43	509.43	427.56
4000	1644.16	1275.02	1016.00	810.91	667.00	555.00	468.26
4500	1693.96	1320.50	1059.08	854.16	708.03	597.53	508.47
5000	1741.84	1366.94	1098.34	895.82	748.01	636.00	549.65

^a Values in parentheses represent vapor pressure expressed in p.s.i. ^b Viscosity expressed in micropoise.

Table III. Coefficients for Polynomial Expressions

Number of Points		Coefficients ^a				Av. Dev.	Std. Dev.	Source of Data
Used	Rejected	A	B	C	D			
Viscosity at Attenuation, η_0								
Ethane								
24	0	-61.82	0.450	-0.401×10^{-3}	0.1812×10^{-6}	0.97 ^b	1.44 ^c	(1, 3, 7, 13)
13	0	25.31	0.0738	-0.132×10^{-3}	0.0634×10^{-6}	0.04	0.06	(1, 3)
Propane								
47	0	23.45	0.0359	0.191×10^{-3}	-0.1090×10^{-6}	0.78	1.06	(1, 2, 5, 10, 12, 13, 15)
25	0	26.14	0.355	0.179×10^{-3}	-0.0984×10^{-6}	0.51	0.09	(1, 2, 12, 13)
n-Butane								
36	0	3.519	0.103	0.090×10^{-3}	-0.0633×10^{-6}	1.19	1.45	(4, 6, 11, 13)
20	0	-34.99	0.283	-0.191×10^{-3}	0.0817×10^{-6}	1.20	1.37	(4, 11)
Residual Viscosity, $\eta - \eta_0$								
Ethane								
317	0	3.171	0.680	-0.0292	0.118×10^{-2}	1.3 ^d	20.3 ^e	(1, 3, 7, 13)
315	2 ^f	2.779	0.794	-0.0365	0.131×10^{-2}	1.7	8.9	
Propane								
417	0	0.008	1.646	-0.0974	0.241×10^{-2}	8.1	25.8	(1, 2, 5, 10, 12, 13, 15)
395	22 ^f	1.870	1.299	-0.0791	0.212×10^{-2}	3.9	23.3	
n-Butane								
283	0	-4.188	1.971	-0.106	0.242×10^{-2}	1.6	17.6	(6, 13)
244	39 ^f	-4.010	1.882	-0.0989	0.229×10^{-2}	0.5	9.4	

^a Coefficient expressed in micropoise. ^b Average deviation expressed in per cent and defined by:

$$s = \left\{ \left[\sum_{i=1}^N [(\eta_0)_e - (\eta_0)_c] / (\eta_0)_e \right] \right\} / N$$

^c Standard deviation defined by: $\sigma = \left[\left\{ \sum_{i=1}^N [(\eta_0)_e - (\eta_0)_c]^2 \right\} / (N - 4) \right]^{1/2}$

^d Average deviation expressed in micropoise and defined by: $s' = \left\{ \sum_{i=1}^N [(\eta - \eta_0)_e - (\eta - \eta_0)_c] \right\} / N$

^e Standard deviation defined by: $\sigma' = \left[\left\{ \sum_{i=1}^N [(\eta - \eta_0)_e - (\eta - \eta_0)_c]^2 \right\} / (N - 1) \right]^{1/2}$

^f Statistically rejected when deviation exceeds $x(\sigma')$ where $x = 2.0$; no rejection factor used for others.

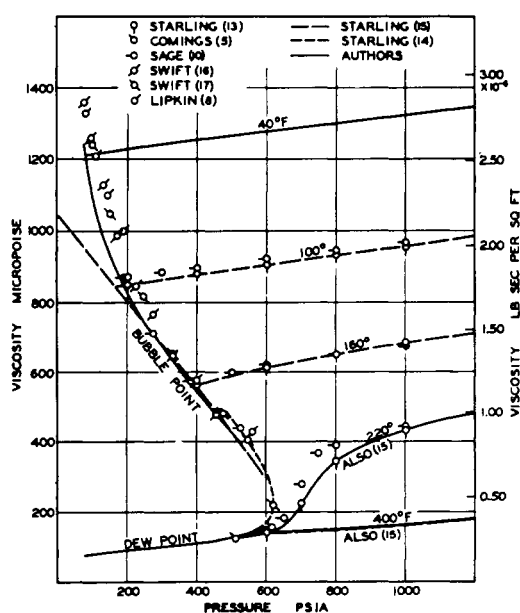


Figure 6. Data from several investigators for propane in the liquid phase

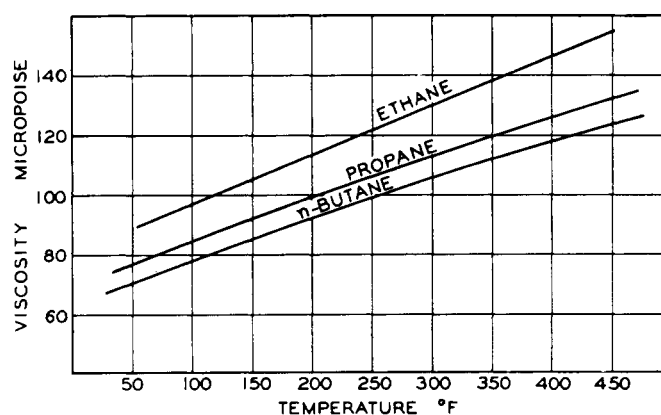


Figure 7. Viscosity of ethane, propane, and n-butane at attenuation

represents the behavior shown in Figure 7. Figure 8 portrays the residual viscosity as a function of specific weight for these same three hydrocarbons. It is apparent that there is a regular progression indicating a smaller increase in residual viscosity with specific weight with the hydrocarbons of higher molecular weight. Again a simple polynomial has been employed in evaluating the residual

viscosity analytically. This polynomial is shown in the following expression:

$$\eta - \eta_0 = A\sigma + B\sigma^2 + C\sigma^3 + D\sigma^4 \quad (2)$$

The standard deviations for ethane, propane, and *n*-butane from the corresponding analytical expressions are submitted in a part of Table III.

If desired, Equations 1 and 2 may be combined to permit the direct evaluation of the viscosity of ethane, propane, and *n*-butane utilizing the coefficients shown in the upper and lower part of Table III.

ACKNOWLEDGMENT

This research was supported by a grant from the Petroleum Research Fund administered by the American Chemical Society. Grateful acknowledgment is hereby made to the donors of this fund. H.H. Reamer contributed to the experimental program, Lorine Faris assisted in the reduction of the data, June Gray prepared the figures, and Grace Fitzsimons assisted in the preparation of the manuscript.

NOMENCLATURE

A, B, C, D = coefficients
N = number of points
s, s' = average deviation (see Table III)
T = thermodynamic temperature, ° R.
x = coefficient of standard deviation
 η = viscosity, micropoise
 Σ = summation operator
 σ = specific weight, lb./cu.ft.
 σ, σ' = standard deviation (see Table III).

Subscripts

c = calculated
e = experimental
o = attenuation

LITERATURE CITED

- (1) Baron, J.D., Roof, J.G., Wells, F.W., *J. Chem. Eng. Data* **4**, 283 (1959).
- (2) Bicher, L.B. Jr., Katz, D.L., *Ind. Eng. Chem.* **35**, 754 (1943).
- (3) Carmichael, L.T., Sage, B.H., *J. Chem. Eng. Data* **8**, 94 (1963).
- (4) *Ibid.*, p. 612.
- (5) Comings, E.W., Mayland, B.J., Egly, R.S., *Univ. Ill. Eng. Exp. Sta. Bull.*, Ser. No. 354, 42, No. 15 (1944).

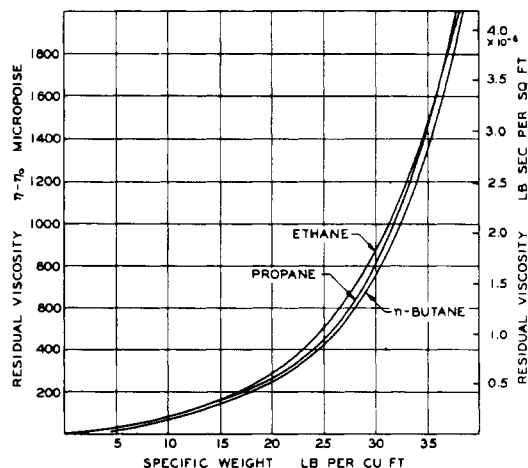


Figure 8. Residual viscosity for ethane, propane, and *n*-butane

- (6) Dolan, J.P., Starling, K.E., Lee, A.L., Eakin, B.E., Ellington, R.T., *J. Chem. Eng. Data* **8**, 396 (1963).
- (7) Eakin, B.E., Starling, K.E., Dolan, J.P., Ellington, R.T., *Ibid.* **7**, 33 (1962).
- (8) Lipkin, M.R., Davison, J.A., Kurtz, S.S., Jr., *Ind. Eng. Chem.* **34**, 976 (1942).
- (9) Reamer, H.H., Cokelet, G.R., Sage, B.H., *Anal. Chem.* **31**, 1422 (1959).
- (10) Sage, B.H., Lacey, W.N., *Ind. Eng. Chem.* **30**, 829 (1938).
- (11) Sage, B.H., Yale, W.D., Lacey, W.N., *Ibid.* **31**, 223 (1939).
- (12) Smith, A.S., Brown, G.G., *Ibid.* **35**, 705 (1943).
- (13) Starling, K.E., Ph.D. thesis, Inst. of Gas Tech., Ill. Inst. of Tech. (1962).
- (14) Starling, K.E., Eakin, B.E., Dolan, J.P., Ellington, R.T., *Prog. Intern. Res. Thermodyn. Transport Properties* Paper Symp. Thermophys. Properties, 2nd, Princeton, N.J., Jan. 24-26, 1962, pp. 530-540, Academic Press, N. Y., 1962.
- (15) Starling, K.E., Eakin, B.E., Ellington, R.T., *A.I.Ch.E.J.* **6**, 438 (1960).
- (16) Swift, G.W., Christy, J.A., Kurata, Fred, *Ibid.* **5**, 98 (1959).
- (17) Swift, G.W., Lohrenz, John, Kurata, Fred, *Ibid.* **6**, 415 (1960).
- (18) Titani, Toshizo, *Bull. Chem. Soc. Japan* **5**, 98 (1930).
- (19) Trautz, Max, Kurz, Friedrich, *Ann. Physik* Ser. 5, **9**, 981 (1931).
- (20) Trautz, Max, Sorg, K.G., *Ibid.* **10**, 81 (1931).

RECEIVED for review December 27, 1963. Accepted March 10, 1964.