(7) Hauser, E.A., Michaels, A.S., *J. Phys. & Colloid Chem.* **52,** 1157 (1948).
(8) Hildebrand, J.H., Scott, R.L., "Regular Solutions," Prentice-Hall, New York, 1962.
(9) Murphy, N F , Lastovica, J E , Fallis, J.G., *Ind. Eng. Chem.* **49,** 1035 (1957).
(10) Newman, M., Hayworth, C.B., Treybal, R.E., *Ibid.*, **41,** 2039 (1949).
(11) Pliskin, I., Ph.D. thesis, New York University, New York, N. Y., 1965.
(12) Pliskin, I., Treybal, R.E., New York University, New York, N. Y., unpublished data, 1965.
(13) Treybal, R.E., "Liquid Extraction," 2nd ed., pp. 131–2, McGraw-Hill, New York, 1963.
(14) Treybal, R.E., Weber, L.D., Daley, J.F., *Ind. Eng. Chem.* **38,** 817 (1946).
(15) Zuidema, E.A., Waters, G.W., *Ind. Eng. Chem., Anal. Ed.* **13,** 312 (1941).

RECEIVED for review July 6, 1965. Accepted October 21, 1965.

Thermal Conductivity of Fluids. Methane

L. T. CARMICHAEL, H. H. REAMER, and B. H. SAGE

Chemical Engineering Laboratory, California Institute of Technology, Pasadena, California

The thermal conductivity of methane was measured at five temperatures between 40° and 340° F. and at pressures between atmospheric and 5000 p.s.i.a. The data were obtained with a spherical thermal conductivity cell and are in good agreement with the measurements of of other investigators at low pressures but show a smaller effect of high pressure upon the thermal conductivity than would be expected from earlier measurements. Within the uncertainty of the present measurements, the thermal conductivity excess is a single-valued function of specific weight. The results are presented in tabular and graphical form.

THE THERMAL conductivity of methane at atmospheric pressure has been studied in some detail. The earlier work of Eucken (*1*) has been supplemented by the more recent measurements of Mann and Dickens (*12*), Johnston and Grilly (*5*), Lambert *et al.* (*9*), and Kannuluik and Donald (*6*). Rather recently, Svehla (*18*) reported values calculated from statistical mechanical considerations and experimental measurements of viscosity at temperatures between −280° and 8540° F. for atmospheric pressure. Schottky (*17*) and Geier and Schafer (*2*) also carried out experimental measurements of thermal conductivity at atmospheric pressure. There is reasonable agreement among these data, and they serve to establish the thermal conductivity of methane satisfactorily at atmospheric pressure. Keyes (*8*) carried out a series of measurements at temperatures between 122° and 572° F. and at pressures as high as 890 p.s.i.a. These data and the measurements by Lenoir and Com-

Table I. Thermal Conductivity of Helium from Several Sources at Atmospheric Pressure

			Thermal Conductivity, B.t.u./(Hr.) (Ft.) (° F.)			
Date	Pressure, P.S.I.A.	Temperature, ° F.	Authors	Keyes[a]	Hilsenrath, Touloukian[b]	Wilson[a]
July 1961	16.3	40	0.08204	0.08273	0.08257	
June 1959	15.0	100	0.08853	0.08864	0.08854	0.0890
July 1960	18.9	100	0.08854			
Aug. 1961	16.6	100	0.08859			
Dec. 1964[c]	18.1	100	0.08824			
Mar. 1962	17.5	130	0.09158	0.09150	0.09135	
Aug. 1962	18.5	130	0.09233			
Dec. 1962	17.2	130	0.09116			
May 1963	15.4	130	0.09138			
Oct. 1963	17.3	130	0.09122			
Jan. 1964[c]	17.4	130	0.09117			
Mar. 1964[c]	17.9	130	0.09071			
May 1964[c]	16.6	130	0.09095			
June 1959	17.7	220	0.09947	0.09960	0.09941	
July 1960	18.8	220	0.09946			
June 1959	15.0	340	0.10941	0.10957	0.10936	
Dec. 1959	15.0	340	0.10966			
July 1960	18.8	340	0.10927			
Aug. 1960	18.0	340	0.10927			
Average deviation[d]				0.00305	0.00291	0.00588

[a] Statistical mechanical calculations and experimental data (*7, 20*).
[b] A critical review (*4*).
[c] Thermal conductivity of methane measurements taken during this time. (January 1964–May 1964, check measurements, December 1964).
[d] Average deviation expressed in fraction and defined by:

$$S' = \frac{\sum_{1}^{N_p} \left| \frac{(k_e)_{av} - k_r}{(k_e)_{av}} \right|}{N_p}$$

ings at 106° F. *(10)* and Lenoir, Junk, and Comings at 127° F. *(11)* at pressures up to nearly 3000 p.s.i.a. appear to be the only measurements that are available concerning the effect of pressure upon the thermal conductivity of methane.

As a result of the paucity of data at high pressures, a series of measurements was made at five temperatures between 40° and 340° F. and at pressures between atmospheric and 5000 p.s.i.a.

EQUIPMENT AND METHOD

Thermal conductivity measurements were carried out in a spherical cell *(14-16)*. Essentially, this apparatus consists of a sphere approximately 3.65 inches in diameter constructed of stainless steel. The exterior surface is gold-plated, and an internal heater of carefully selected geometry serves to provide the energy scource. This sphere is located within a spherical cavity, the interior of which is also gold-plated. The radial transport path between the inner sphere and outer shell is approximately 0.020 inch, and nearly equal fluxes were encountered throughout the path. Thermocouples were employed to establish the temperature of the inner spherical surface and that of the inner surface of the outer cavity. Appropriate corrections were made for the location of the thermocouples within the stainless steel of the inner sphere and the outer cavity *(16)*. Furthermore, the dimensions of the gap of the transport path were established by direct measurements with appropriate corrections for the effects of pressure and temperature upon the length of the path *(16)*.

The over-all performance of the instrument has been checked periodically by measurements of the thermal conductivity of helium and argon *(3, 4, 7)* at atmospheric pressure. As an illustration of the reproducibility of the thermal conductivity cell over a period of time, values of the measured thermal conductivity of helium at atmospheric pressure and a comparison with reported data *(4, 7, 20)* are given in Table I. The comparison is satisfactory with an over-all average deviation of 0.32%. The variation in the measured values with time is larger than might be expected. A portion of the variation may be due to diffusion of traces of hydrocarbons of intermediate molecular weight through the closely fitting joints of the inner surface of the hemisphere from the small free volumes in the region of the unsupported area seal *(14-16)*. Small traces of impurity introduce a marked change in the apparent thermal conductivity of helium.

Table II presents several values of the thermal conductivity of helium at elevated pressures *(10, 19, 20)*. Apparently, there is only limited information available concerning the behavior of helium at elevated pressure. In the comparison, in order to separate the effect of pressure from the variation in absolute values at atmospheric pressure, the quotient of the reported value at pressure to the extrapolated value at attenuation has been recorded.

The agreement of the values shown in Tables I and II, which was established from measurements of the dimensions of the instrument and the energy added to the inner sphere as well as from the temperature differences between the two surfaces, is an indication of the reproducibility and accuracy of the instrument now in use. The current measurements on methane were carried out between January 1964 and May 1964, and these data were bracketed with the measurements on helium, as indicated in Tables I and II.

Table II. Effect of Pressure on Thermal Conductivity of Helium

Pressure, P.S.I.A.	Thermal Conductivity Ratio [a] Authors	Vodar[b]	Wilson[c]	Lenoir[d]
	At 100° F.			At 109° F.
1239	1.0147	1.0190	1.0189	1.0305
2583	1.0350	1.0394	1.0396	1.0636
4046	1.0563	1.0614[e]	1.0618[e]	1.0995[e]

[a] Quotient of thermal conductivity at pressure to the extrapolated value at attenuation.
[b] Reference *(19)*.
[c] Calculated from statistical mechanical considerations *(20)*.
[d] Reference *(10)*.
[e] Values of thermal conductivity at this pressure extrapolated from data at lower pressures.

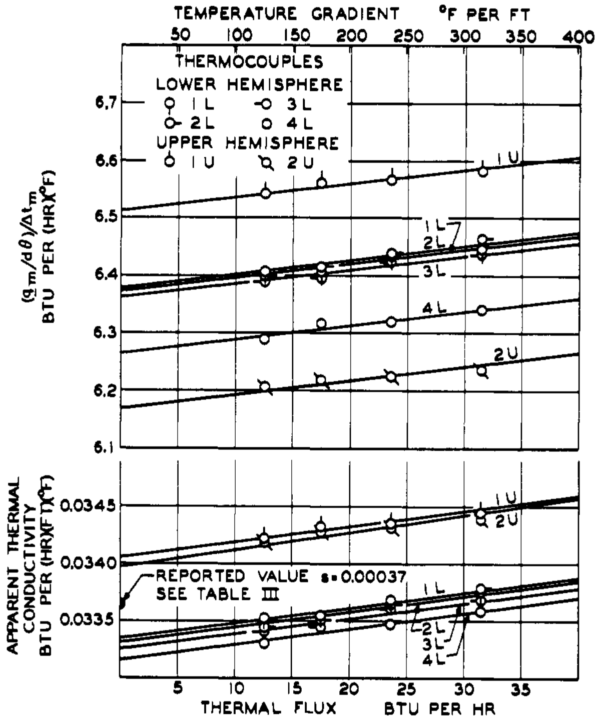

Figure 1. Effect of thermal flux upon apparent thermal conductivity at 2098 p.s.i.a. and 280° F.

MATERIALS

The methane employed in this investigation was obtained from the Texas Co. and, in the as-received condition, involved approximately 0.0003 mole fraction of hydrocarbons other than methane and approximately 0.0020 mole fraction of nitrogen. The gas at a pressure in excess of 500 p.s.i.a. was passed through a metal trap held at the temperature of solid carbon dioxide and acetone and passed over calcium chloride, activated charcoal, potassium hydroxide, and anhydrous calcium sulfate.

EXPERIMENTAL RESULTS

To illustrate the way in which the therml conductivities were established, Figure 1 shows the influence of thermal flux upon the quantity $(q_m/d\Theta)/\Delta t_m$. As the temperature gradient is increased, there is a gradual increase in this quantity. Furthermore, there is a significant difference between the values obtained among the several thermocouples. These are experimental data upon which no corrections have been made. These data were obtained at a pressure of 2098 p.s.i.a. and a temperature of 280° F.

The apparent thermal conductivity for each of the thermocouples is shown in the lower part of Figure 1. These thermocouples have been corrected for their position in the shell and in the inner sphere. These corrections have brought the data for the lower hemisphere in good agreement, as has been done for the upper hemisphere. There exists a small lack of symmetry between the upper and lower hemispheres as a result of the effect of pressure on the relative position of the centers of the inner spherical cavity and the internal sphere which amounts to as much as 0.001 inch at higher pressures. An analysis of the transport phenomena indicates that a simple linear correction suffices to relate the data for the upper and lower hemispheres.

The use of a simple averaging process does not introduce more than 0.1% uncertainty in the reported value of the thermal conductivity corresponding to the behavior of the instrument at zero thermal flux. In Figure 1, the slope of the line is the same for both the upper and lower hemispheres.

The experimental results are presented in Table III. The standard error of estimate of the points obtained with the six different thermocouples from the least-squares fit of the straight lines shown in Figure 1 has been indicated in this table. The value of the thermal conductivity calculated from these measurements, as has been described, is reported along with the standard deviation of this value which is reported from the area-weighted averages of the values obtained from the indications of the six thermocouples. The means of evaluating each of the measures of uncertainty has been indicated. As a result of a slightly smaller effect of pressure upon the thermal conductivity than had been reported by others (*8*, *10*, *11*), a supplemental pair of measurements was made after recalibration with helium at atomospheric and elevated pressure. Satisfactory agreement between the earlier and more recent data was obtained.

Table III. Experimental Conditions and Results

Pressure, P.S.I.A.	Number Flux Values	Maximum Flux, B.t.u./Hr.	Number of Points	Gradient [a], ° F.$^{-1}$	Standard Error of Estimate[b], (B.t.u./Hr.)/(° F.)	Thermal Conductivity, B.t.u./(Hr.)(Ft.)(° F.)	Standard Deviation,[c] B.t.u./(Hr.)(Ft.)(° F.)
			40° F.				
17	4	27.03	24	0.00301	0.00358	0.018050	0.000333
992	4	29.42	24	0.00143	0.01320	0.021894	0.000132
2017	4	35.29	24	0.00267	0.01047	0.029150	0.000089
3742	4	39.26	24	0.00099	0.01293	0.040662	0.000594
4974	4	36.38	24	0.00064	0.00547	0.046836	0.001067
			100° F.				
17	4	27.37	24	0.00289	0.00335	0.020372	0.000119
952	4	31.03	24	0.00219	0.00400	0.023199	0.000088
1960	4	29.55	24	0.00109	0.00592	0.028145	0.000093
3934	3	29.30	18	0.00174	0.00612	0.038144	0.000692
4977	4	36.70	24	0.00127	0.00609	0.042693	0.001088
17[d]	4	23.48	24	0.00304	0.00536	0.020264	0.000086
3998[d]	4	27.22	24	0.00254	0.00612	0.038317	0.000605
			220° F.				
17	4	22.89	24	0.00399	0.00659	0.025803	0.000090
1027	4	27.20	24	0.00220	0.00494	0.028112	0.000039
1979	3	28.47	18	0.00166	0.01390	0.031272	0.000344
3911	4	33.55	24	0.00419 [e] 0.00182 [f]	0.01742	0.037337	0.000894
4778	4	34.23	24	0.00228 [e] 0.00039 [f]	0.00392	0.040186	0.001202
			280° F.				
18	4	30.29	24	0.00331 [e] 0.00211 [f]	0.00450	0.029201	0.000089
1018	4	29.93	24	0.00240 [e] 0.00100 [f]	0.00673	0.031147	0.000196
2098	4	31.49	24	0.00239	0.00577	0.033641	0.000370
3928	4	34.32	24	0.00230	0.00643	0.038509	0.000857
5124	4	38.92	24	0.00208 [e] 0.00103 [f]	0.00635	0.041844	0.001349
			340° F.				
18	4	28.99	24	0.00089	0.00959	0.032214	0.000081
952	4	30.23	24	0.00329 [e] 0.00119 [f]	0.00592	0.033802	0.000270
2034	4	34.37	24	0.00214 [e] 0.00064 [f]	0.00410	0.036015	0.000417
3995	4	33.54	24	0.00235 [e] 0.00124 [f]	0.00578	0.040458	0.000901
5114	4	33.33	24	0.00071	0.00729	0.043155	0.001125

[a] Average value of gradient over all thermocouple measurements defined as:

$$\frac{d[(q_m/d\theta)/\Delta t_m]}{d(q_m/d\theta)}$$

[b] Standard error of estimate:

$$\sigma = \left[\frac{\sum_{1}^{N_p}\left[\left(\frac{q_m/d\theta}{\Delta t_m}\right)_e - \left(\frac{q_m/d\theta}{\Delta t_m}\right)_s\right]^2}{N_p - 1}\right]^{1/2}$$

[c] Standard deviation from area weighted average of the indications of the six thermocouples:

$$s = \left[\frac{\sum_{1}^{N_p}(k'_{\text{av}} - k')^2}{N_p}\right]^{1/2}$$

[d] Check measurements.

[e] Average value of gradient of thermocouples in lower hemisphere.

[f] Average value of gradient of thermocouples in upper hemisphere.

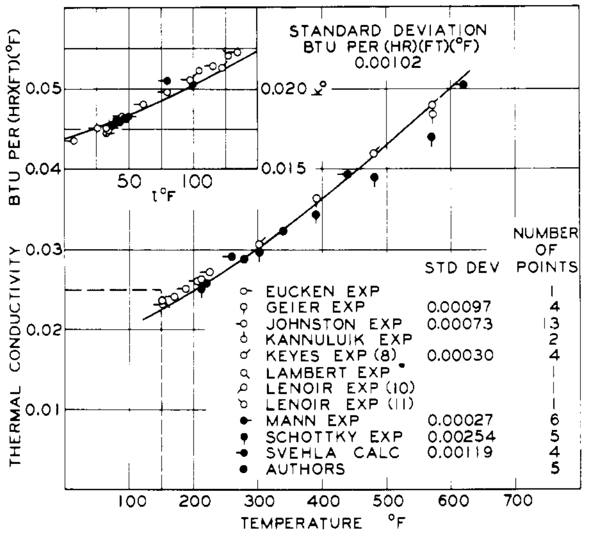

Figure 2. Thermal conductivity of methane at attenuation

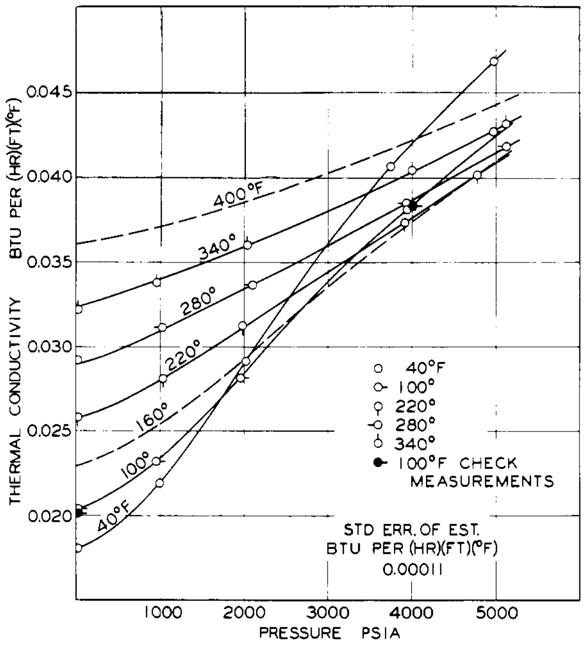

Figure 3. Thermal conductivity of methane

Approximately 25 measurements of the apparent thermal conductivity were made for each of the points shown in Table III. Consideration of Table III indicates a larger standard deviation with an increase in pressure which is to be expected as a result of the diverging concentricity of the inner sphere and the outer cavity. However, as was indicated earlier, a simple averaging process does not in itself introduce more than 0.1% added uncertainty in the results. The average deviation shown in Table III takes into account the correction for the lack of concentricity prior to the evaluation of the deviations and is relatively independent of pressure.

The effect of temperature upon the thermal conductivity of methane at pressures near attenuation is shown in Figure 2. Experimental values obtained at pressures near atmospheric were corrected to attenuation from a knowledge of the rate of change of the thermal conductivity with pressure. The measurements at temperatures below 150° F. have been shown on an enlarged scale because of the greater amount of experimental data available in this region. The current data at temperatures from 100° to 220° F. yield values slightly lower than those reported by Johnston and Grilly (*5*) and slightly below the low-temperature data of Svehla (*18*). However, an extrapolation of the current data is in good agreement with the experimental values of Keyes (*8*) and the calculated values of Svehla (*18*). The latter's data at low temperatures are somewhat higher than the values obtained by a number of investigators. The standard deviation for several investigators (*2*, *5*, *8*, *12*, *17*, *18*) has been included on the diagram together with the number of data points involved. The over-all standard deviation (*1*, *2*, *5*, *6*, *8*-*12*, *17*, *18*) corresponds to an average deviation of approximately 2.6% of the average value of the thermal conductivity. The smooth curve shown in Figure 2 was based upon the five

Table IV. Thermal Conductivity of Methane

Pressure, P.S.I.A.	Temperature, ° F.						
	40	100	160[a]	220	280	340	400[b]
14.7	0.01804[c]	0.02036	0.02292	0.02578	0.02894	0.03238	0.03606
200	0.01850	0.02074	0.02327	0.02609	0.02924	0.03265	0.03625
400	0.01912	0.02125	0.02369	0.02648	0.02962	0.03299	0.03644
600	0.01990	0.02188	0.02423	0.02693	0.03001	0.03331	0.03666
800	0.02083	0.02258	0.02479	0.02745	0.03047	0.03368	0.03686
1000	0.02193	0.02336	0.02543	0.02802	0.03095	0.03403	0.03710
1500	0.02526	0.02580	0.02724	0.02955	0.03220	0.03492	0.03779
2000	0.02903	0.02845	0.02925	0.03119	0.03339	0.03595	0.03854
2500	0.03272	0.03128	0.03150	0.03286	0.03468	0.03697	0.03934
3000	0.03618	0.03390	0.03361	0.03443	0.03602	0.03806	0.04027
3500	0.03928	0.03622	0.03549	0.03602	0.03739	0.03921	0.04119
4000	0.04200	0.03842	0.03741	0.03764	0.03870	0.04036	0.04220
4500	0.04452	0.04046	0.03927	0.03928	0.04007	0.04153	0.04320
5000	0.04695	0.04245	0.04107	0.04090	0.04145	0.04273	0.04427
σ[d]	0.00000	0.00017		0.00007	0.00016	0.00013	

[a] Values interpolated with respect to temperature.
[b] Values extrapolated with respect to temperature.
[c] Thermal conductivity expressed in B.t.u./(hr.) (ft.) (° F.).
[d] Standard error of estimate, σ, expressed in B.t.u./(hr.) (ft.) (° F.) and defined as:

$$\sigma = \left[\frac{\sum_{1}^{N_p} (k_e - k_s)^2}{N_p - 1}\right]^{1/2}$$

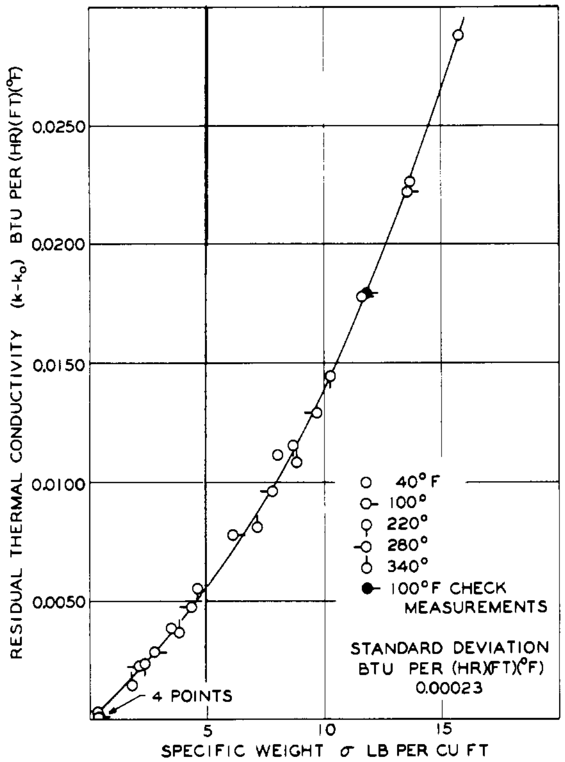

Figure 4. Residual thermal conductivity of methane

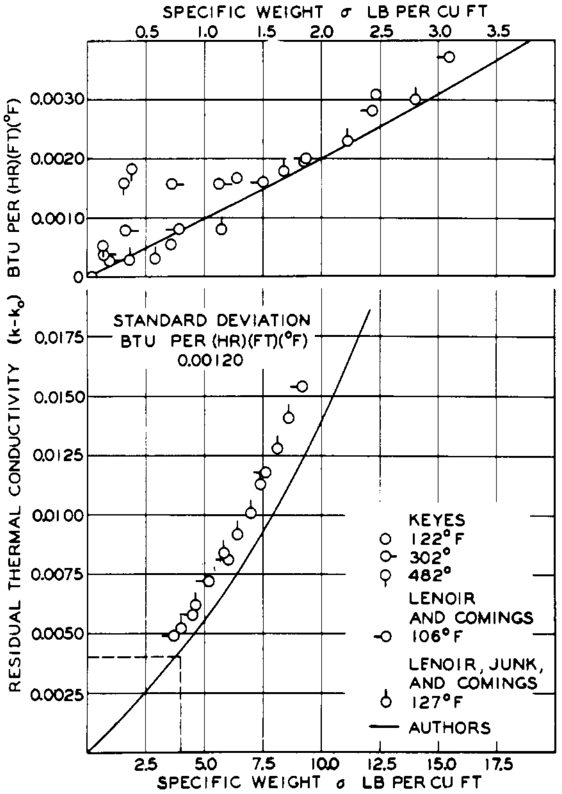

Figure 5. Comparison of measurements from several investigators

current measurements plus the higher-temperature experimental data of Keyes (*8*). The recently calculated values reported by Svehla (*18*) were included for comparison.

Figure 3 depicts the experimental measurements obtained in the current investigation. The behavior interpolated for 160° F. and extrapolated for 400° F. has been shown as dashed curves. The agreement of the two states studied after the completion of this investigation and re-evaluation of the behavior with helium have been shown as "check measurements." The standard error of estimate was 0.00011 B.t.u./(hr.) (ft.) (°F.). This corresponds to a relative standard error of approximately 0.3% based upon the average thermal conductivity. Such agreement is within the uncertainty of measurement.

The influence of specific weight of methane upon the residual thermal conductivity, also known as the thermal conductivity excess, is shown in Figure 4. In arriving at the information shown, a study of the volumetric behavior of methane (*13*) was employed to establish the specific weight as a function of pressure and temperature. The information shown in Figure 4 may readily be described by a polynomial of the following form:

$$k - k_0 = A\sigma + B\sigma^2 + C\sigma^3 + D\sigma^4 \qquad (1)$$

The constants were established by conventional regression analysis and yielded a standard deviation of 0.00023 B.t.u./(hr.) (ft.) (°F.). The standard deviation is defined as:

$$\sigma' = \left[\frac{\sum_{1}^{N_p} [(k - k_0)_e - (k - k_0)]^2}{N_p - N_c} \right]^{1/2} \qquad (2)$$

The values of the constants of Equation 1 are as follows:

$A = 960.904 \times 10^{-6}$ $\qquad C = 2.95427 \times 10^{-6}$

$B = 17.2505 \times 10^{-6}$ $\qquad D = -0.0357371 \times 10^{-6}$

The check measurements at 100° F. were in good agreement with the earlier data.

Figure 5 shows the measurements of other investigators (*8, 10, 11*) that have been made at pressures significantly above that of the atmosphere. The measurements of Lenoir and Comings (*10*) and of Lenoir, Junk, and Comings (*11*) yield a higher value of residual thermal conductivity at a given specific weight than do the current measurements. The measurements by Keyes (*8*) do not extend to a sufficiently high pressure to permit worthwhile comparison. The behavior at low pressures has been shown on an enlarged scale in the upper part of Figure 5. As would be expected, the variation among the several investigators is relatively large. The standard deviation of all of the experimental data shown in Figure 5 was 0.00120 B.t.u./(hr.) (ft.) (°F.).

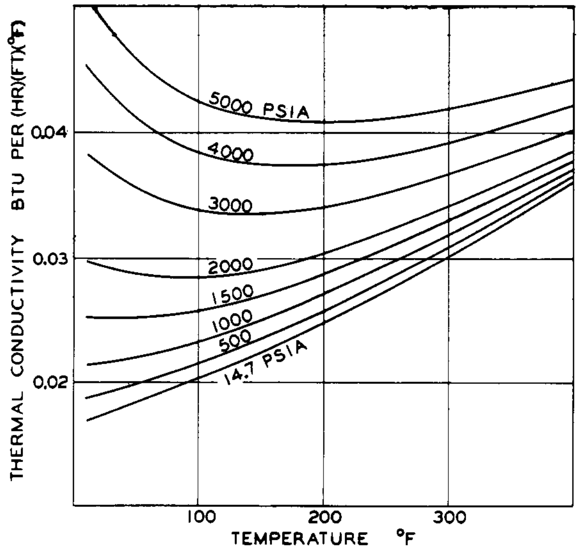

Figure 6. Effect of temperature upon the thermal conductivity of methane

It is not clear why the current measurements yield a smaller effect of pressure on the thermal conductivity than the data of Lenoir *et al.* (*10*, *11*) which were obtained with an entirely different type of instrument. This perhaps serves to illustrate the need for measurements of molecular transport characteristics to be carried out with two or more widely different types of instruments before great confidence can be placed in the results. The authors have found helium a satisfactory reference standard at atmospheric pressure and are attempting to develop a background of information concerning the small effect of pressure upon the thermal conductivity of helium in order that such information may be used for direct comparison of the behavior of thermal conductivity equipment. As was indicated, the measurements on helium are set forth in Tables I and II.

The influence of temperature on the thermal conductivity of methane throughout the range of pressures covered in this investigation is shown in Figure 6. The behavior is typical of that found for hydrocarbons at temperatures well above the critial temperature of the compound in question. Smooth values of the thermal conductivity of methane are reported in Table IV, including interpolated values for 160° F. and extrapolated values for 400° F. The standard error of estimate of these measurements from the experimental data reported in this manuscript is indicated for each temperature where measurements were made. The over-all standard error of estimate for the smooth data shown in Table IV was 0.00011 B.t.u./(hr.) (ft.) (°F.).

ACKNOWLEDGMENT

This experimental work was carried out through the support of the National Science Foundation under Grant GP-2357. Joan Jacobs reduced the experimental results while Virginia Berry carried out the regression analysis to establish the coefficients of Equation 1. G. N. Richter contributed earlier to this program by an evaluation of the effects of the position of thermocouples and a lack of concentricity of the inner sphere and outer cavity. Grace Fitzsimons contributed to the preparation of the manuscript.

NOMENCLATURE

A, B, C, D = coefficients
d = differential operator
k = thermal conductivity, B.t.u./(hr.) (ft.) (° F.)
k_0 = thermal conductivity at attenuation, B.t.u./(hr.) (ft.) (° F.)
k' = thermal conductivity uncorrected for effect of pressure on instrument, B.t.u./(hr.) (ft.) (° F.)
N_C = number of constants
N_P = number of points
$q_m/d\theta$ = measured thermal flux, B.t.u./hr.
s = standard deviation defined in Table III
s' = average deviation expressed in fraction and defined in Table I
Δt_m = measured temperature difference, ° F.
σ = specific weight, lb./cu. ft.
σ = standard error of estimate defined in Tables III and IV
σ' = standard deviation defined in Equation 2
Σ = summation operator
θ = time, hr.

Subscripts

av = average
c = calculated
e = experimental
r = reference
s = smoothed

LITERATURE CITED

(1) Eucken, A. von, *Physik. Z.* **14,** 324 (1913).
(2) Geier, H. von, Schafer, K., *Allgem. Waermetech.* **10,** 70 (1961).
(3) Hilsenath, J., *et al.*, "Tables of Thermal Properties of Gases," *Natl. Bur. Std.* (*U.S.*), *Circ.* No. **564** (1955).
(4) Hilsenrath, J., Touloukian, Y.S., *Trans. Am. Soc. Mech. Engrs.* **76,** 967 (1954).
(5) Johnston, H.L., Grilly, E.R., *J. Chem. Phys.* **14,** 233 (1946).
(6) Kannuluik, W.B., Donald, H.B., *Austral. J. Sci. Res.* **3A,** 417 (1950).
(7) Keyes, F.G., *Trans. Am. Soc. Mech. Engrs.* **73,** 589 (1951).
(8) Keyes, F.G., *Ibid.*, **76,** 809 (1954).
(9) Lambert, J.D., Cotton, K.J., Pailthorpe, M.W., Robinson, A.M., Scrivins, J., Vale, W.R.F., Young, R.M., *Proc. Roy. Soc.* (*London*) **A231,** 280 (1955).
(10) Lenoir, J.M., Comings, E.W., *Chem. Eng. Progr.* **47,** 223 (1951).
(11) Lenoir, J.M., Junk, W.A., Comings, E.W., *Ibid.*, **49,** 539 (1953).
(12) Mann, W.B., Dickins, B.G., *Proc. Roy. Soc.* (*London*) **134-A,** 77 (1931–32).
(13) Olds, R.H., Reamer, H.H., Sage, B.H., Lacey, W.N., *Ind. Eng. Chem.* **35,** 922 (1943).
(14) Richter, G.N., Sage, B.H., *Chem. Eng. Data Ser.* **2,** 61 (1957).
(15) Richter, G.N., Sage, B.H., J. CHEM. ENG. DATA **4,** 36 (1959).
(16) *Ibid.* **8,** 221 (1963).
(17) Schottky, W.F. von, *Z. Elektrochem.* **56,** 889 (1952).
(18) Svehla, R.A., *NASA Tech. Rept.* **R-132** (1962).
(19) Vodar, B., "Heat Conductivity of Helium," report written for Electric Boat Div. of Gen. Dyn. Corp., 1959.
(20) Wilson, M.P., General Dynamics Corp., GA-1355, TID-4500, 15th Ed., January 1960.

RECEIVED for review April 30, 1965. Accepted September 27, 1965. This paper was accepted as a contribution to this journal by R.L. Pigford, Editor of *Ind. Eng. Chem. Fundamentals*.