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Thermal Conductivity of Five Hydrocarbons Along the Saturation Line

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The paper presents the results of new, absolute measurements of the thermal conductivity of five alkanes as a function of temperature at their saturation vapor pressure. The alkanes studied include the four normal alkanes n-octane, n-nonane, n-undecane, and n-tetradecane as well as the branched alkane, iso-octane. The results, which extend over the temperature range 282–373 K, have an estimated uncertainty limit of 1.5% deriving mainly from a correction for the effects of radiation absorption in the measurement process. The results are employed to generate effective core volumes for the fluids studied which may be employed to predict the density dependence of the thermal conductivity by means of an existing correlation.

KEY WORDS: alkanes; hydrocarbons; liquids; thermal conductivity.

1. INTRODUCTION

In a series of earlier papers [1–5], we have described the design and development of a transient hot-wire instrument for accurate measurements of the thermal conductivity of liquids along their saturation line. In continuation of this work, we present here results of measurements upon a series of four normal alkanes (octane, nonane, undecane, and tetradecane) and one branched alkane (iso-octane) in the temperature range 282–373 K. For the first time in our work it has proved possible to correct the experimental data for the effects of radiative heat transport so that the present results enjoy a higher level of accuracy than hitherto, and prove to be consistent with independent measurements at elevated pressures [6, 7].

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In the case of the normal alkanes, the present results, in conjunction with the correlation scheme proposed by Menashe et al. [8], allow the prediction of the density dependence of the thermal conductivity over a wider range of temperature than was possible before. However, the branched alkane, iso-octane, has been found to have a substantially lower thermal conductivity than its normal isomer. For this reason, and because the correlation scheme has not yet been extended to branched isomers of alkanes, iso-octane must currently be omitted from the calculation scheme.

2. EXPERIMENTAL PROCEDURE

The instrument employed for the measurements has been described in detail elsewhere [1, 5]. In particular, the most recent design of thermal conductivity cells, in which a weight is employed to tension the wires, has been described by the authors [5]. The characteristics of the various wires employed during the present series of measurements are listed in Table I. The liquid samples were supplied by B. D. H. Chemicals Ltd. (n-octane, n-nonane, n-undecane, and n-tetradecane), and Merck (iso-octane), and they were distilled under vacuum and degassed before use. The purities of the hydrocarbons were confirmed to be in excess of 99% by gas chromatographic analysis and by refractive index measurements. The results of the latter are collected in Table II where they are compared with standard values [9]. The mean extinction coefficients for radiation absorption, κ_a [10], which are required to correct the data for radiative heat transport, have been determined under ambient conditions, and their values are also included in Table II. It should be noted that where these results can be compared with the values of Menashe and Wakeham [6, 11], they are some 30–40% higher. It is thought that this difference is a measure of the

Table I. Characteristics of the Hot-Wire Cells

Series I: n-octane, n-nonane	
Wire radius, a	$5.49 \pm 0.08\ \mu m$
Long wire length, l_1	143.98 ± 0.01 mm
Short wire length, l_2	37.85 ± 0.01 mm
Cell radius, b	5.0 ± 0.1 mm
Series II: iso-octane, n-undecane, n-tetradecane	
Wire radius, a	$5.49 \pm 0.08\ \mu m$
Long wire length, l_1	145.47 ± 0.01 mm
Short wire length, l_2	39.97 ± 0.01 mm
Cell radius, b	5.0 ± 0.1 mm

Table II. Optical Characteristics of the Liquids

Liquid	Measured refractive index, n (25 °C)	Standard refractive index,[a] n (25 °C)	Extinction coefficient, κ_a (m^{-1})
n-octane	1.39503	1.39511[b]	1544
i-octane	1.38892	1.38893	1898
n-nonane	1.40346	1.40338	1574
n-undecane	1.41502	1.41507	1684
n-tetradecane	1.42675	1.42685	1742

[a] From ref. [9].

[b] Extrapolated.

precision with which this difficult measurement can be made by standard techniques and contributes to our estimate of the uncertainty in the thermal conductivity data [6, 10, 11].

The thermal conductivity measurements were carried out in the temperature range 282–333 K for n-octane. For the remaining systems the highest temperature reached was approximately 370 K. The lowest temperature attained for n-tetradecane was limited by the need to avoid solidification. The densities and heat capacities of the fluids, required for the application of small corrections in the reduction of the experimental data, have been obtained from available tabulations [12, 13], supplemented by suitable estimation procedures when necessary [14]. In most cases a transient temperature rise of between 4 and 6 K was employed in the measurements, but for iso-octane, a somewhat greater value of 6 to 9 K was used.

2.1. Working Equations

The majority of the working equations for the transient hot-wire method are now well established, and details may be found elsewhere [15–17]. However, in the present instrument the temperature rise of the wires is somewhat greater than that assumed in the original analysis of the method [15], so that it is necessary to consider afresh one of the corrections to be applied in the reduction of the data. Specifically, we examine the direct effect of the temperature dependence of the fluid thermophysical properties.

Following the analysis of Healy et al. [15], we account for the temperature dependence of the fluid properties by means of linear perturbations about their equilibrium values so that

$$\lambda(T, \rho) = \lambda(T_0, \rho_0)(1 + \chi \Delta T) \tag{1}$$

and

$$\rho C_p = \rho_0 C_{p0}(1 + \phi \Delta T) \tag{2}$$

in which the coefficient χ accounts for both the direct influence of temperature on the thermal conductivity and the indirect influence through its dependence on the changing density. Solving the Fourier heat conduction equation iteratively for these perturbations to the standard problem, we obtain the equation [15]

$$\ln\left[\frac{4\kappa(T_0, \rho_0)}{a^2 C} t\right] = \frac{-q}{4\pi\lambda_0}(\chi - \phi)\ln 4 + \frac{4\pi\lambda_0}{q}\Delta T + \frac{2\pi\lambda_0}{q}\chi(\Delta T)^2 \tag{3}$$

for the evolution of the temperature rise of the wire. This quadratic relationship indicates that the line ΔT versus $\ln t$ will exhibit a small, but usually imperceptible curvature.

In practice [1], a set of experimental data points $(\Delta T_i, \ln t_i)$ for $i = 1$ to n is fitted by a least squares method to the equation

$$\ln\left[\frac{4\kappa(T_0, \rho_0)}{a^2 C} t\right] = A + B\Delta T \tag{4}$$

in which

$$B = \frac{4\pi\lambda(T_r, \rho_r)}{q} \tag{5}$$

Consequently, in order to determine the appropriate value of the reference temperature T_r we must impose the condition that Eq. (4) provides the best representation of a set of data described by Eq. (3) in a least squares sense. This condition leads to the result that

$$B = \frac{4\pi\lambda(T_0, \rho_0)}{q}\{1 + \chi\delta T_2^*\} \tag{6}$$

in which we retain the notation of ref. [15], and where

$$\delta T_2^* = \frac{1}{2}\left\{\frac{\sum_{i=1}^{n}(\Delta T_i)^3 - \frac{1}{n}\sum_{i=1}^{n}(\Delta T_i)^2 \sum_{i=1}^{n}\Delta T_i}{\sum_{i=1}^{n}(\Delta T_i)^2 - \frac{1}{n}\left(\sum \Delta T_i\right)^2}\right\} \tag{7}$$

A comparison of Eq. (6) with Eq. (5) and use of Eq. (1) allows us to identify immediately the reference temperature T_r as

$$T_r = T_0 + \delta T_2^* \tag{8}$$

This result for the reference temperature differs algebrically from that given by Healy et al. [15],

$$T_r = T_0 + \tfrac{1}{2}(\Delta T_1 + \Delta T_n) \tag{9}$$

but reduces to it in the special case when the data points are uniformly distributed along the ΔT axis ($\Delta T_{i+1} - \Delta T_i = \text{constant}$). In general, this condition cannot be met exactly in practice, and there is a small numerical difference between the two reference temperatures. In typical measurements the difference may amount to as much as 0.1 K for a total temperature rise of 4 K and to 0.2 K for a total temperature rise of 8 K.

For many measurements, because the thermal conductivity is a weak function of temperature ($(1/\lambda) d\lambda/dT \simeq 2 \times 10^{-3}\ \mathrm{K}^{-1}$), the difference in reference temperature is insignificant. However, in special cases when the temperature dependence of the thermal conductivity is stronger, the difference can be significant, particularly if large temperature rises are employed. Consequently, to safeguard the highest precision in our measurements, we have consistently adopted Eqs. (7) and (8) for the evaluation of the reference temperature.

The thermal conductivity $\lambda(T_r, \rho_r)$ obtained by the preceding analysis includes a contribution from the radiative heat flux inevitably present in the measurement process. Menashe and Wakeham [10] have carried out a numerical analysis of the contribution of this radiative heat flux to the apparent thermal conductivity for absorbing fluids and have proposed a method whereby the data may be corrected to yield radiation-free values. We have applied their procedure to our data and give results for both the apparent and radiation-free thermal conductivities.

3. RESULTS

Tables III to VII list the thermal conductivity data for the five alkanes, n-octane, iso-octane, n-nonane, n-undecane, and n-tetradecane, respectively, as a function of temperature at the saturation vapor pressure. The tables include the apparent thermal conductivity $\lambda_{\text{app}}(T, \rho)$ as well as the radiation-free values $\lambda(T, \rho)$. The precision of the thermal conductivity measurements is estimated to be ±0.2%. However, the correction to the

Table III. Thermal Conductivity of n-Octane at Its Saturation Vapor Pressure, P_s

Temperature T (K)	Apparent thermal conductivity, $\lambda_{\text{app}}(T, P_s)$ $(\text{mW} \cdot \text{m}^{-1} \cdot \text{K}^{-1})$	Thermal conductivity $\lambda(T, P_s)$ $(\text{mW} \cdot \text{m}^{-1} \cdot \text{K}^{-1})$
282.09	134.7	132.4
285.50	133.5	131.1
285.50	133.5	131.2
289.86	131.5	129.1
293.74	130.7	128.3
293.93	130.4	128.0
298.46	129.0	126.5
301.96	128.1	125.5
305.80	126.0	123.4
309.93	125.0	122.3
311.44	124.3	121.6
311.55	124.5	121.9
315.44	122.9	120.2
317.27	122.5	119.7
317.35	122.5	119.7
317.38	122.7	119.9
321.14	120.9	118.1
323.56	120.3	117.5
327.44	119.0	116.1
333.24	117.5	114.6
336.81	116.0	113.1

Table IV. Thermal Conductivity of Iso-octane at Its Saturation Vapor Pressure, P_s

Temperature T (K)	Apparent thermal conductivity $\lambda_{\text{app}}(T, P_s)$ $(\text{mW} \cdot \text{m}^{-1} \cdot \text{K}^{-1})$	Thermal conductivity $\lambda(T, P_s)$ $(\text{mW} \cdot \text{m}^{-1} \cdot \text{K}^{-1})$
292.45	97.28	94.68
303.60	94.94	92.18
311.26	93.12	90.25
311.71	92.98	90.11
320.14	90.89	87.91
320.16	90.76	87.77
329.35	88.74	85.65
336.52	86.88	83.71
337.08	86.90	83.73
346.33	84.53	81.27
346.36	84.46	81.21
357.79	82.06	78.70
357.84	82.17	78.80
359.36	81.68	78.30
359.47	81.61	78.24

Table V. Thermal Conductivity of n-Nonane and Its Saturation Vapor Pressure, P_s

Temperature T (K)	Apparent thermal conductivity $\lambda_{\mathrm{app}}(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	Thermal conductivity $\lambda(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$
281.98	136.8	134.6
285.87	135.5	133.2
292.02	133.4	131.0
292.48	133.5	131.1
295.89	132.5	130.0
296.16	131.8	129.3
296.17	132.1	129.7
297.63	131.6	129.1
297.79	131.8	129.3
301.61	130.1	127.6
303.47	129.9	127.3
303.65	129.6	127.0
303.69	129.5	126.9
303.69	129.7	127.0
307.36	128.6	125.9
309.31	128.0	125.3
309.45	128.0	125.3
313.12	127.1	124.3
315.63	126.2	123.4
321.57	124.4	121.5
324.92	123.7	120.7
327.78	122.3	119.3
327.81	122.8	119.8
331.53	121.2	118.2
331.56	121.3	118.2
336.72	119.8	116.7
336.73	119.8	116.8
340.73	118.6	115.5
340.80	118.6	115.4
348.47	116.4	113.1
352.17	115.5	112.2
357.99	113.7	110.3
361.60	112.7	109.3

experimental data necessary to account for the effects of radiation absorption amounts to as much as 4%. It is estimated [6, 10, 11] that the uncertainty in the value of this correction, arising both from errors in the determination of the mean extinction coefficient and the numerical evaluation of the correction, may be as much as 20%. Consequently, we estimate that the overall uncertainty of our experimental data is, at its worst, ± 1.5%.

For the purposes of interpolation within the present data we have represented the thermal conductivity of each fluid along the saturation line

Table VI. Thermal Conductivity of n-Undecane at Its Saturation Vapor Pressure, P_s

Temperature T (K)	Apparent thermal conductivity $\lambda_{\mathrm{app}}(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	Thermal conductivity $\lambda(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$
284.61	137.2	134.9
289.66	135.8	133.3
294.72	134.5	131.9
299.78	133.2	130.5
299.79	132.9	130.2
304.10	131.8	129.0
304.31	131.8	129.0
304.38	131.7	128.9
309.54	130.1	127.2
309.56	130.3	127.4
314.01	128.7	125.7
314.06	128.9	125.9
318.70	127.7	124.6
318.82	128.0	124.9
320.77	127.0	123.9
325.02	125.9	122.8
325.12	125.7	122.5
327.36	125.2	122.0
330.95	124.3	121.1
330.97	124.8	121.6
336.84	122.7	119.3
339.21	122.1	118.7
339.25	122.4	119.0
339.28	121.8	118.5
340.51	121.9	118.5
340.58	122.0	118.6
347.94	120.2	116.6
347.95	119.5	116.0
354.22	118.3	114.7
354.25	118.3	114.6
354.25	118.1	114.5
359.57	116.8	113.1
359.58	116.9	113.2
364.38	115.5	111.7
364.88	115.6	118.8
370.18	113.8	110.0
370.18	114.0	110.1
370.41	113.6	109.8
373.36	112.8	108.9

Table VII. Thermal Conductivity of n-Tetradecane at Its Saturation Vapor Pressure, P_s

Temperature T (K)	Apparent thermal conductivity $\lambda_{\mathrm{app}}(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	Thermal conductivity $\lambda(T, P_s)$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$
301.30	140.4	137.6
301.39	140.1	137.3
309.51	138.1	135.0
316.17	136.5	133.3
321.07	135.1	131.8
324.79	133.6	130.3
331.20	132.2	128.7
331.57	131.6	128.1
334.08	131.2	127.7
336.85	130.6	127.0
336.86	130.4	126.8
342.90	129.4	125.7
342.99	129.2	125.5
353.84	125.8	121.9
357.09	125.3	121.3
357.59	125.1	121.1
357.61	125.2	121.2
366.06	122.5	118.4
366.46	122.8	118.7
366.78	122.7	118.5
367.31	122.7	118.5

by means of the correlation

$$\lambda(T, P_s) = \lambda_0 + C(T - 273.15) \tag{10}$$

The coefficients which provide the best representation of the data are included in Table VIII. Among the normal alkanes the values of both

Table VIII. Coefficients of the Correlation of the Thermal Conductivity of the Alkanes Along the Saturation Line

Liquid	λ_0 $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	C $(\mathrm{mW \cdot m^{-1} \cdot K^{-2}})$
n-octane	135.38	−0.3533
iso-octane	99.547	−0.2474
n-nonane	136.87	−0.3159
n-undecane	137.88	−0.2880
n-tetradecane	145.48	−0.2890

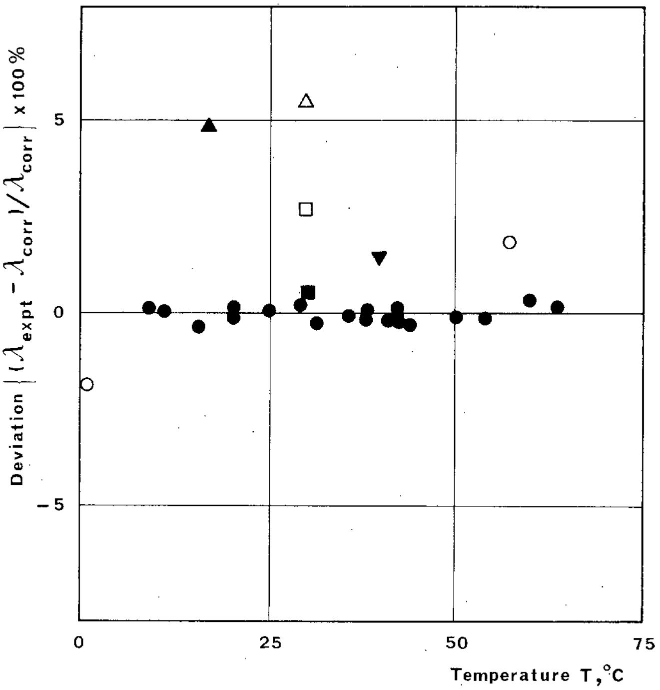

Fig. 1. Deviations of the thermal conductivity data for n-octane from the correlation of the present results. ● Present work; ○ [18]; □ [19]; △ [20]; ▲ [21]; ■ [22]; ▼ [23].

coefficients show a systematic trend with increasing chain length. Figures 1 to 5 contain plots of the deviations of the present experimental data from the correlation for the five hydrocarbons; the deviations do not exceed ±0.3%, whereas the standard deviations are less than 0.2%. The same figures include the results of earlier measurements [6, 18–31]. The only previous sets of data which have been corrected for the contribution from radiation are those reported by Menashe and Wakeham [6] for n-nonane and n-undecane. Their measurements were performed at elevated pressures and did not extend to the saturation line. Nevertheless, the values obtained by extrapolation of their data using the correlation procedure given by Menashe et al. [8] are consistent with the present data. For n-nonane the earlier results lie approximately 1% below the present correlation, whereas for n-undecane they lie approximately 0.7% above. In view of the large extrapolation of the earlier results involved in this comparison and the estimated uncertainties in both sets of data, the agreement must be regarded as satisfactory. The results of the remaining measurements reported

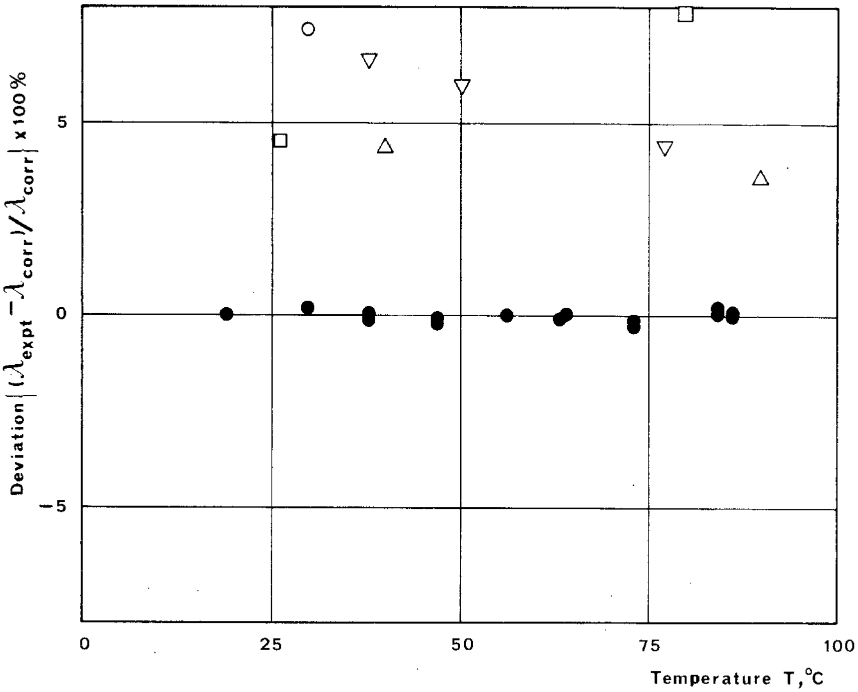

Fig. 2. Deviations of the thermal conductivity data for iso-octane from the correlation of the present results. ● Present work; □ [24]; △ [23]; ▽ [31]; ○ [26].

in the literature have not been corrected for the effects of radiation, and they generally deviate substantially from the present correlation, although individual data points sometimes lie within the claimed mutual uncertainty. The results reported here are to be preferred owing to their high accuracy.

4. DISCUSSION

The first observation which follows from the experimental data concerns the large difference between the thermal conductivity of the two isomers of octane. Normal octane has a thermal conductivity which is very similar to that of other normal alkanes, whereas the value for iso-octane is some 30% lower. This observation is somewhat surprising because other properties of the two isomers are much less different. For example, the heat capacities differ by no more than 6%, the molar volumes by at most 2%, and the viscosities [32] by no more than 10%. These facts suggest that the difference in the thermal conductivity is entirely due to the reduced ability

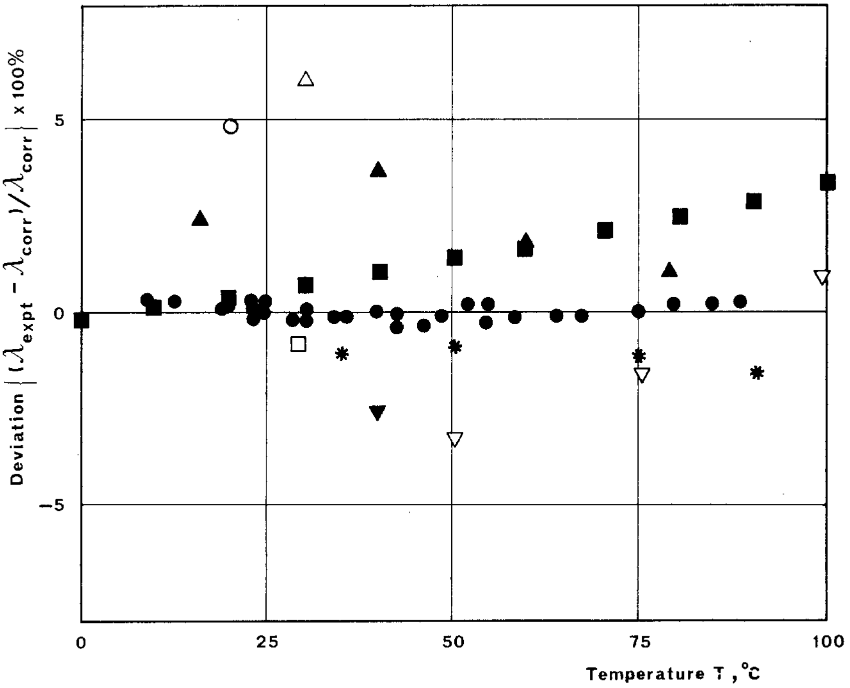

Fig. 3. Deviations of the thermal conductivity data for n-nonane from the correlation of the present results. ● Present work; □ [19]; △ [20]; ▽ [27]; ■ [22]; ▼ [28]; ▲ [21]; ○ [29]; * [6].

of the branched alkane to transport energy and not to changes in the free volume within the liquid. In the absence of a suitable theory of thermal conductivity in polyatomic liquids, it is not fruitful to speculate further on the origins of this difference at this stage, but it will be interesting to examine whether a similar difference is observed between isomers of other alkanes.

The correlation scheme proposed by Menashe et al. [8] for the density dependence of the thermal conductivity of normal alkanes is based upon an empirical extension of Dymond's treatment of the Enskog theory for dense fluids [33, 34]. According to this correlation scheme, the quantity

$$\lambda' = \frac{1.936 \times 10^7 \lambda(V,T) V^{2/3} (M/RT)^{1/2}}{[1 + 0.352 C_{v,\text{int}}/R]} \tag{11}$$

is a function of (V/V_0) only. In this equation V is the molar volume of the liquid, M the molar mass, R the universal gas constant, and $C_{v,\text{int}}$ the contribution of internal degrees of freedom to the ideal gas heat capacity at

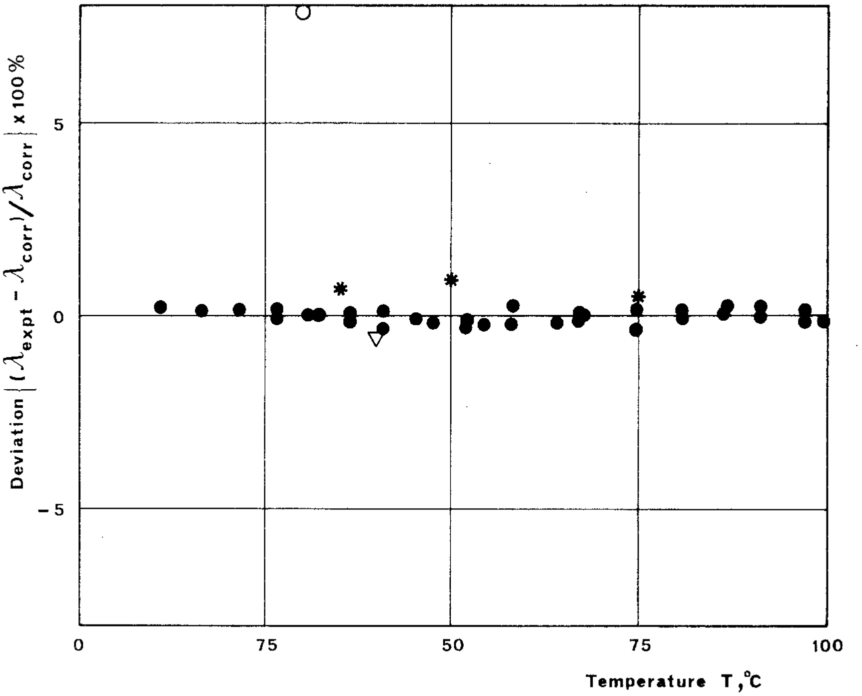

Fig. 4. Deviations of the thermal conductivity data for n-undecane from the correlation of the present results. ● Present work; ○ [20]; ▽ [28]; * [6].

constant volume. The quantity V_0 is a characteristic molar volume for the fluid which is a weak function of temperature. It has been shown [8] that λ' is a universal function of (V/V_0) for several normal alkanes, and this function has been correlated by means of the equation

$$\ln \lambda' = 2.8724 - 2.2735 \ln(V/V_0) \qquad (12)$$

which describes the available experimental data with a deviation of less than ±1% [8, 35]. The form of Eqs. (11) and (12) shows that just one measurement of the thermal conductivity of a normal alkane at one density and temperature is sufficient to determine the characteristic volume V_0 at that temperature. Consequently, it is then possible to predict the density dependence of the thermal conductivity along the isotherm. It has already been demonstrated that this procedure yields predicted thermal conductivities with an error of only ± 1% [8]. We have therefore employed the present data for the thermal conductivity along the saturation line of each normal alkane to determine $V_0(T)$, and the values are listed in Table IX. For this

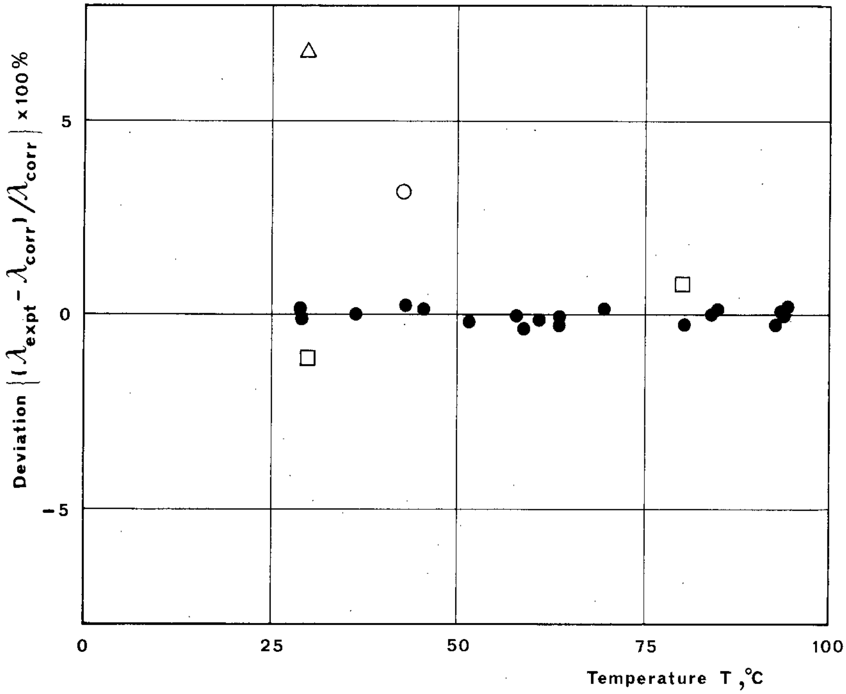

Fig. 5. Deviations of the thermal conductivity data for n-tetradecane from the correlation of the present results. ● Present work; △ [20]; □ [30]; ○ [31].

Table IX. Characteristic Molar Volumes, V_0, for the Normal Alkanes

Temperature T(K)	n-Octane	n-Nonane	n-Undecane	n-Tetradecane
	V_0 (10^{-5}m^3 · mol^{-1})			
280	10.61	11.62		
290	10.41	11.45	13.84	
300	10.24	11.28	13.61	17.36
310	10.07	11.12	13.40	17.08
320	9.906	10.96	13.19	16.81
330	9.755	10.82	13.00	16.55
340		10.68	12.81	16.31
350		10.55	12.64	16.08
360		10.40	12.48	15.86
370			12.32	

purpose we have employed the density data given by Doolittle [36] and Rossini [12].

The values of V_0 given in Table IX together with Eqs. (11) and (12) permit the evaluation of the thermal conductivity of the normal alkanes studied in this work over a wide range of thermodynamic states. It is expected that the accuracy of these predicted values will be comparable with that of direct measurements.

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NOTE ADDED IN PROOF

New results on radiation theory show that the radiation-conduction interaction term is negligible for the liquids discussed in this paper. It is now apparent that the most accurate values of the Thermal conductivity are those listed in Tables III–VII under the heading ‘Apparent thermal conductivity,’ and the overall uncertainty of the data is now less than 1%.

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