σ	$\theta_0'(0)$	$\theta_1'(0)$	$\theta_2'(0)$
0·7	−0·29268	−0·66364	0·62688
1	−0·33206	−0·69432	0·65658
10	−0·72814	−1·01761	1·12404
20	−0·925	−1·180	1·380
30	−1·0506	−1·2847	1·55885
100	−1·5718	−1·7206	2·302
300	−2·267	−2·3011	3·2946
500	−2·68	−2·652	3·8987
1000	−3·3870	−3·23875	4·9065
1500	−3·885	−3·655	5·625
2000	−4·2668	−3·9730	6·185
5000	−5·790	−5·2434	8·3733

where $\theta_0'(0)$, $\theta_1'(0)$ and $\theta_2'(0)$ have the same signification as in [2]. They are obtained by differenciation with $\eta = 0$ of the relative concentrations (or temperatures) $\theta_0(\eta)$, $\theta_1(\eta)$, $\theta_2(\eta)$ which appear in the series proposed to describe the concentration (or temperature) profiles in the boundary layer:

$$\theta = \theta_0(\eta) + \xi \cdot \theta_1(\eta) + \xi^2 \cdot \theta_2(\eta) + \cdots \qquad (7)$$

These numerical results permit the calculation of the local mass (or heat) transfer coefficient to a cylinder, k_{cylinder}, for small values of the curvature parameter ξ, in an extended range of σ. This coefficient has the following expression:

$$k_{\text{cylinder}} = -\frac{\alpha}{R \cdot \xi}\{\theta_0'(0) + \xi \cdot \theta_1'(0) + \xi^2 \cdot \theta_2'(0) + \cdots\} \quad (8)$$

in which α symbolizes the diffusion coefficient of the diffusing component or the thermal conductivity of the fluid.

When $R \to \infty$, equation (8) gives the local mass (or heat) transfer coefficient for a laminar boundary layer over a flat plate:

$$k_{\text{f.p.}} = -\theta_0'(0) \cdot \alpha \cdot \left(\frac{U}{\nu x}\right)^{1/2}. \qquad (9)$$

It can be seen that the tabulated values of $\theta_0'(0)$ agree very well with the asymptotic solution obtained by Cess [7]:

$$\theta_0'(0) = -0{\cdot}3388 \cdot \sigma^{1/3}. \qquad (10)$$

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USE OF THERMAL COMPARATOR METHOD FOR THERMAL CONDUCTIVITY MEASUREMENTS ON LIQUIDS: VALUES FOR THREE ORGANIC SERIES: NORMAL ALCOHOLS, ACIDS AND SATURATED HYDROCARBONS

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NOMENCLATURE

C, degree Celsius;
K, Kelvin;
m, metre;
n, number of carbon atoms;
W, watt.

INTRODUCTION

FOLLOWING the development by the senior author of thermal comparator methods for thermal conductivity determinations on liquids [1–4], few further such applications appear to have been reported, only that of Shroff [5] having been noticed.

The purpose of the present account is to demonstrate by a few practical examples the value of this simple method as further developed at the Thermophysical Properties Research Center* [6, 7], in helping to resolve uncertainties in liquid thermal conductivity data.

NEW MEASUREMENTS RELATING TO TESTS ON LIQUIDS

For all thermal comparator tests on liquids the liquids are totally enclosed to avoid evaporation and associated local cooling, and contact is made on a thin stretched membrane which covers or forms the base of the cell containing the liquid and makes good contact with the liquid. For the present work, 5 cells, each $1\frac{1}{4}$ in. dia. and $\frac{7}{8}$ in. deep are mounted side by side in an aluminum plate and successive contacts on the top of each were made by a probe mounted on a form of balance.

Three series of organic liquids have been investigated for fairly comprehensive ranges of carbon number, *n*. These are the alcohols, $C_nH_{2n+1}OH$, for $n = 1$–9, and 11 and 12, the saturated hydrocarbons, C_nH_{2n+2} for $n = 6$–17 and the acids $C_nH_{2n}O_2$ for $n = 1$–9.

A preliminary test series of measurements made with water filling each of the five cells showed the results for each cell to be closely similar. By arranging for the five cells to contain two liquids of known thermal conductivity and three different test liquids, tests could readily be made under closely comparable conditions for the five cells. Plots of thermal comparator reading against the observed differences between the probe and sample temperatures were fitted by the best straight line. The thermal comparator readings for each liquid were then derived for a chosen difference of 16°C. Ethanol and toluene were mainly used as the reference liquids, their thermal conductivities at 30°C were assumed to be $0{\cdot}165_1$ and $0{\cdot}133_2$ W m^{-1} K^{-1} respectively in agreement with the TPRC derived values [8]. It is of interest to note that a 1970 Russian handbook [9] gives closely similar values of $0{\cdot}165_5$ W m^{-1} K^{-1} for ethanol and $0{\cdot}132_6$ W m^{-1} K^{-1} for toluene. On the other hand Poltz and Jugel [10] point out that since toluene is a liquid that absorbs heat radiation poorly the radiative component causes the effective thermal conductivity to be thickness dependent. They find the true thermal conductivity to be $0{\cdot}127_7$ W m^{-1} K^{-1} at 30°C for zero thickness, the effective combined conduction and radiation value being $0{\cdot}135_3$ for an infinite thickness and $0{\cdot}133_3$ for a thickness of about 2 mm. It is thought that the present assumed values are accurate to ±2 per cent. These uncertainties, however, will have a reduced influence on the relative values obtained for other liquids by the present comparative method. In the latter the accuracy will depend on such other factors as liquid purity, diaphragm reproducibility, temperature inaccuracies and the digital nature of the recording instrument. The over-all accuracy is probably of the order of ±3–4 per cent.

RESULTS AND DISCUSSION

(a) *The saturated (aliphatic) hydrocarbons*, C_nH_{2n+2}

The saturated hydrocarbons, C_nH_{2n+2}, belong to a series for which many thermal conductivity determinations have been made, at least for the members having $n \geqslant 5$ for which the normal boiling point is above 30°C. The available data for 30°C taken from 23 sources are reproduced in Fig. 1 where thermal conductivity values are plotted against *n*. The figure's main purpose is to show both the large departures of a few determinations and the concentration of the larger proportion within a band which indicates a definite tendency for the thermal conductivity at 30°C for this series of liquids to increase with *n* at a rate which decreases as *n* increases. The band width extends about ±6, ±4, ±2·5 and ±2 per cent from the means for *n*-values of 5, 9, 13 and 18 respectively. Curves 18–20 represent the results of assessments and semi-empirical correlations that have been proposed by workers in Great Britain [32], France [33], and the United States of America [8], respectively. They are seen to agree reasonably well.

The inset portion of Fig. 1 shows that at 100°C data of [32] and [33] agree to well within their respective accuracy limits of ±5 and ±4 per cent. The earlier TPRC data [8] also agree, but would appear to deviate if extrapolated above their present limit of $n = 10$. At 200°C the differences are somewhat greater but the mean values come well within the stated accuracies. The subsequent data of Rastorguyev *et al.* [34] are definitely lower.

Special comment should be made of the 30°C values for $n = 9$, 10 and 12 attributed to Filippov [18] and later reproduced by Tsederberg [19], since both authors reference these as from data by Smith [13]. Clearly there is some inconsistency as Smith's actual values are reproduced in Curve 3 and are about 14 per cent greater.

The values now obtained are plotted in Fig. 1 as large squares and are included in the caption. They are seen to be in reasonable accord with the above-mentioned Curves 18–20, particularly for the higher values of *n*. For $n = 6$–8 the present values are 4–5 per cent greater than those of Curves 18 and 19 so lie just within their expected accuracy limits.

It is of interest to note that for n-octadecane, which has the largest value of *n* included in the main figure, the experimental values of [25, 26, 29] and the derived values of [32, 33] all lie within ±2 per cent of the value indicated by a mean curve through the present results. This curve is shown as a broken line.

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* Information on the availability of this instrument may be obtained from the McClure Park Corp., 2595 Yeager Road, W. Lafayette, Indiana 47906.

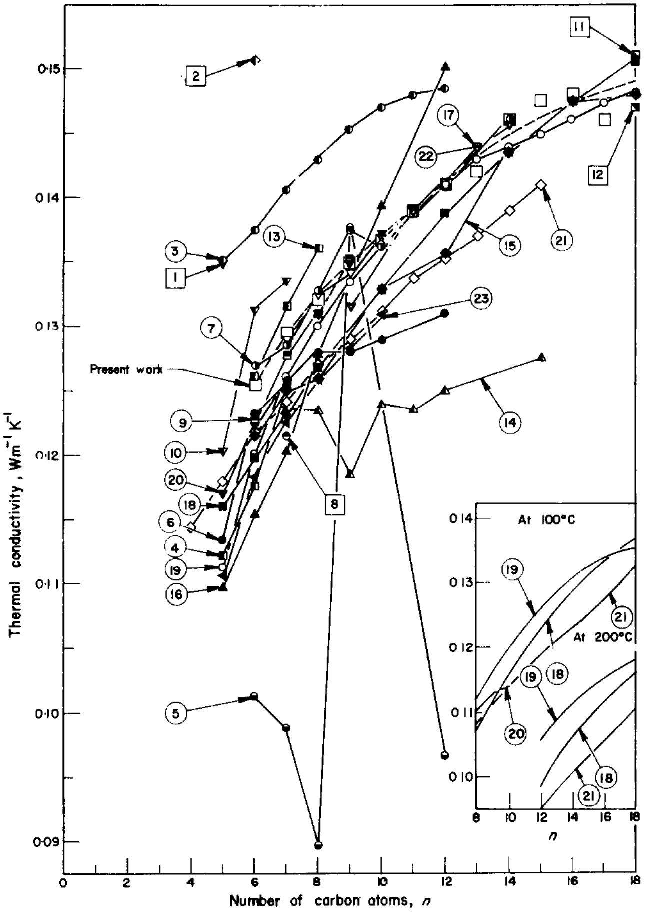

FIG. 1. Thermal conductivity of saturated (aliphatic) hydrocarbons, C_nH_{2n+2}, at 30°C plotted against n.

1 Bridgman [11], 2 Shiba [12], 3 Smith [13], 4 Riedel [14], 5 Skrynnikova [15] (data as reported by Jamieson and Tudhope [16]), 6 Filippov [17, 18] also Tsederberg [19], 7 Sakiadis and Coates [20, 21], 8 Briggs [22], 9 Frontasev and Gusakov [23], 10 Vilím [24], 11 Powell and Challoner [25], 12 Ziebland and Patient [26], 13 Golubev and Naziev [27], 14 Akhmedov [28], 15 Mukhametzianov, Usmanov and Tarzimanov [29], 16 Jobst [30], 17 Abas-Zade and Guseinov [31], 18 Reynard and Walton [32], 19 Missenard [33], 20 Liley [8], 21 Rastorguyev, Bogatov and Grigor'yev [34], 22 Abas-Zade, Guseinov and Efendiyev [35], 23 Brykov, Mukhametzianov and Usmanov [36].

Large squares represent present results (0·125$_5$, 0·129$_5$, 0·132, 0·135, 0·136$_5$, 0·139, 0·141, 0·142, 0·146, 0·147$_5$, 0·148 and 0·146 W m^{-1} K^{-1} for n = 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16 and 17 respectively) and broken line the smooth-fitting curve.

The nature of the agreement between the present results and the recommended values for this much-studied series is regarded as supporting the reliability of the thermal comparator method for these measurements.

(b) *The normal acid series*, $C_nH_{2n}O_2$

Figure 2 contains thermal conductivity values at 30°C for the normal acid series plotted against the number of carbon atoms. The experimental information on this series is relatively sparse, the only measurements covering a good proportion of the early members of this series being those of Jobst [30]. The other values reproduced are from Tsederberg [19] and Missenard [33]. Those of the former are based on Russian data and on the early measurements by Weber [37]. These values now appear to decrease too rapidly as n increases from 2 to 5.

The present values are shown as squares in Fig. 2 and are given in the caption. For the measurements on formic acid ($n = 1$), glycerin was included as a reference liquid, and the thermal conductivity of formic acid proved to be above 0·288 W m^{-1} K^{-1}, assumed for the glycerin used. The present values of 0·298 W m^{-1} K^{-1} is in fair agreement with the Riedel [14] value of 0·296 W m^{-1} K^{-1} at 20°C but exceeds the values of Filippov [17] and Vargaftik [38, 39] by 9 and 16 per cent respectively.

For most of the values of n from 2 to 9 the present results

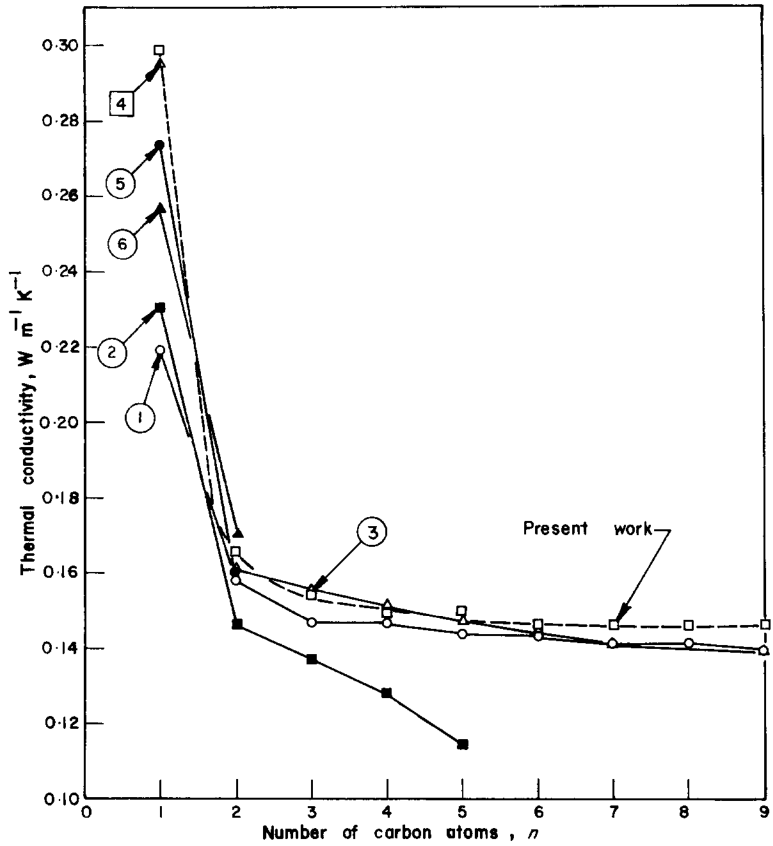

FIG. 2. Thermal conductivity of the normal acid series $C_nH_{2n}O_2$ at 30°C plotted against n.
1 Jobst [30], 2 Tsederberg [19], 3 Missenard [33], 4 Riedel [14], 5 Filippov [17], 6 Vargaftik [38, 39].
Squares represent present results (0·298, 0·165, 0·153$_5$, 0·149, 0·149$_5$, 0·146, 0·146, 0·146 and 0·146 W m^{-1} K^{-1}, for n = 1, 2, 3, 4, 5, 6, 7, 8 and 9 respectively) and broken line the smooth-fitting curve.

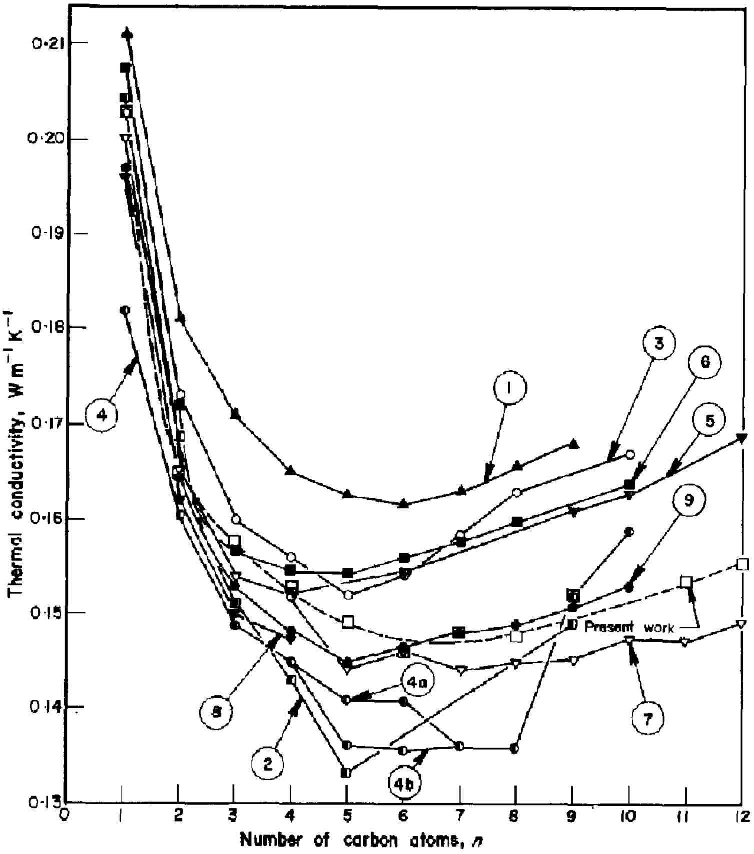

FIG. 3. Thermal conductivity of the alcohols, $C_nH_{2n+1}OH$, at 30°C plotted against n.

1 Daniloff [40], 2 Filippov [17], 3 Sakiadis and Coates [20], 4 Jobst [30], 4a from Fig. 2, 4b from Fig. 7 of Ref. [30], 5 Mukhametzianov, Tarzimanov and Usmanov [41], 6 Tufeu, LeNeindre, Bury and Johannin from Fig. 13 of Ref. [42], 7 Shroff [5], 8 Poltz and Jugel [43], 9 Missenard [33].

Large squares represent present results (0·203, 0·165—Ref. [8] value)—$0{\cdot}157_5$, 0·153, 0·149, 0·146, 0·148, $0{\cdot}147_5$, 0·152, 0·153 and $0{\cdot}155_5$ W m^{-1} K^{-1} for $n = 1$, 2, 3, 4, 5, 6, 7, 8, 9, 11 and 12 respectively), and broken line the smooth-fitting curve.

are in fair accord with the Missenard [33] data based on the experimental values of Jobst [30]. A value of 0·146 W m^{-1} K^{-1} was found for values of $n = 6$–9.

(c) *The normal alcohol series*, $C_nH_{2n-1}OH$

Figure 3 is a similar plot for normal alcohol series, except that, in view of the number of measurements made for some of the earlier members, only those data are included for which the same workers gave values for several members of the series. Even so, differences of from 16 to 22 per cent are apparent; also the value of n for which the thermal conductivity at 30°C is a minimum appears to be uncertain The data by Daniloff [40] are now regarded as being high, since he used data by Bridgman [11] for methanol and n-butanol to calibrate an apparatus of similar design. The present results again happen to be in fair accord with the values derived from another equation due to Missenard [33].

Jobst [30] indicated two different values for both $n = 5$ and $n = 6$ in two figures of his paper. Both are included in

Fig. 3. His values in this region are seen to be the lowest, apart from one value by Filippov [17], but an unusually sharp rise is noted as n increases from 8 to 10.

It is interesting to observe, that of the experimental results for the higher values of n, the present values agree best with those of Shroff [5] who also employed a thermal comparator method. The differences do not exceed 5 per cent and could include sample differences as well as experimental uncertainties. These last were estimated as ±3·1 per cent by Shroff and ±3–4 per cent in the present work. Both sets of thermal comparator determinations have indicated a minimum thermal conductivity to occur when n is about 7, whereas others, including [20, 41, 42] found the minimum to occur for n values of about 4 or 5.

CONCLUSIONS

For each of the three series of organic liquids the thermal conductivity values furnished by these measurements have been found to vary in a fairly regular manner with increase in the number of carbon atoms. The smooth curves fitting the present thermal conductivity results are thought to give true values to within about ±3 per cent accuracy, the small departures from a smooth curve being attributed to experimental limitations and possibly to sample impurities.

The thermal comparator method has been proved to offer a useful and rapid means for checking some of the large thermal conductivity departures that exist for liquids. It is a method that could be added to advantage in many laboratories employing other thermal conductivity methods. For instance, to quote just one example, its use would have enabled Jobst [30] to confirm whether the thermal conductivity of his sample of n-decanol was actually some 17 per cent greater than that of his sample of n-octanol. Confirmation would have supported his original measurements, but have indicated impurities in the liquids used, whereas non-conformation would have indicated the need for careful re-examination of the original determinations.

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