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Measurements and Correlation of the Thermal Conductivity of Liquid *n*-Paraffin Hydrocarbons and Their Binary and Ternary Mixtures

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The thermal conductivity of four pure normal paraffin hydrocarbons (C_{11}, C_{14}, C_{15}, C_{16}) and binary and ternary mixtures of three *n*-paraffin hydrocarbons (C_7, C_{11}, C_{16}) have been measured in the temperature range from about 20 to 90°C at atmospheric pressure. Measurements have been performed with the aid of a fully automated transient hot-wire instrument. The accuracy of the reported data is estimated to be ± 1.0 to $\pm 1.5\%$. A new simple and practical equation, which can calculate the thermal conductivity of pure *n*-paraffin hydrocarbons ($4 \leqslant n \leqslant 16$) with the uncertainty of $\pm 1\%$, has been correlated in terms of temperature and number of carbon atoms based on the present results including some other reliable data. Also, a mixing rule for the mixtures of *n*-paraffin hydrocarbons was proposed and was adequately confirmed by the present results within the experimental error.

KEY WORDS: alkanes; hydrocarbons; liquids; mixing rule; thermal conductivity; transient hot-wire method.

1. INTRODUCTION

Liquid paraffin hydrocarbons (alkanes; C_nH_{2n+2}) and their mixtures have widely been employed as heat carriers or thermal energy storage materials, since these substances are chemically stable and easy to handle. Besides, one can easily obtain desired melting and boiling points by choosing their appropriate carbon numbers. If someone needs the thermal conductivity values of the liquid state for these substances, so far two ways to estimate them seem to exist. First, estimation methods, which might be represented by the method of Robbins and Kingrea [1], can be used as a rough

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estimation. Although these estimation methods can be applied to many kinds of substances, the equations include many thermophysical properties such as heat capacity, critical temperature, and heat of vaporization, whose reliable values often are not easy to find. Moreover, it is not unusual for these methods to produce some 10 or 20% error. Second, we can use very accurate correlations for individual liquid hydrocarbons as a function of temperature and pressure which are based on recent measurements using the transient hot-wire method [2–7]. These correlations may be reliable individually, however, from the viewpoint of practical applications, they are not interrelated with each other and cannot be used for mixtures.

Taking into account the facts mentioned above, we have been attempting to develop a generalized correlation for the thermal conductivity of liquid paraffin hydrocarbons, which has to be practically useful, simple, and reasonably accurate, on the basis of precise measurements performed in a systematic way. In order to carry out such a series of measurements, then, a fully automated apparatus without any reduction of accuracy is desirable for use. A companion paper [8], therefore, is devoted to the development of a new fully automated transient hot-wire apparatus.

The present paper describes a practical and accurate correlation for the thermal conductivity of liquid *n*-paraffin hydrocarbons and their mixtures as well as a set of systematically obtained experimental results for these substances.

2. EXPERIMENTAL PROCEDURE

The instrument for the measurements has been described in detail in a companion paper [8]. In the instrument particular emphasis is placed on the following features. (i) The time of onset of natural convection can automatically be detected for each run, then the thermal conductivity can be calculated using the data between appropriate time intervals. (ii) All sequential switchings and resistance adjustment procedures can be done in an automatic way. (iii) Temperature control for the thermostatic bath is automated as well. After reaching some preset temperature, the instrument starts to take measurements. The characteristics of a typical hot wire used in the present series of measurements are listed in Table I.

Table I. Characteristics of a Typical Hot Wire

Radius of wire	$30 \pm 0.3\ \mu m$
Length of wire	74.92 ± 0.01 mm
Resistance–temperature relation of wire	$R(T) = 10.3782(1 + 3.9477 \times 10^{-3} \cdot T - 5.50 \times 10^{-7} \cdot T^2)$

The liquid samples employed in the present measurements (*n*-heptane *n*-C_7H_{16}, *n*-undecane *n*-$C_{11}H_{24}$, *n*-tetradecane *n*-$C_{14}H_{30}$, *n*-pentadecane *n*-$C_{15}H_{32}$, and *n*-hexadecane *n*-$C_{16}H_{34}$) are reagent grade, have a stated purity of not less than 99%, and were used without further purification processes. The mixtures were prepared gravimetrically by means of a precision balance.

The resistance of the wire was calibrated *in situ* against the temperature before a series of thermal coductivity measurements. The temperature was measured with the aid of a standard platinum resistance thermometer which was calibrated on an IPTS-68.

The thermal conductivity measurements were carried out in the temperature range from about 20 to 90°C at atmospheric pressure for pure substances. For the mixtures the highest temperature was approximately 70°C. The current through the wire was automatically chosen so as to attain a transient temperature rise of about 1 K at 3 s after the initiation of heating.

3. RESULTS AND CORRELATION FOR PURE *n*-PARAFFIN HYDROCARBONS

Tables II to V list the thermal conductivity data for the four normal paraffin hydrocarbons, *n*-undecane, *n*-tetradecane, *n*-pentadecane, and *n*-hexadecane, respectively, as a function of temperature at atmospheric pressure. These values are the averages of two to five measurements whose reproducibility was less than ±0.7%. And the overall accuracy of our experimental data is estimated to be ±1.0%. Detailed assessment of the

Table II. Thermal Conductivity of *n*-Undecane (*n*-$C_{11}H_{24}$) at Atmospheric Pressure

Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
19.3	0.1353
30.9	0.1317
40.8	0.1292
49.6	0.1279
60.3	0.1259
70.3	0.1244
80.8	0.1221
90.7	0.1195

Table III. Thermal Conductivity of *n*-Tetradecane (n-$C_{14}H_{30}$) at Atmospheric Pressure

Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
11.1	0.1432
21.4	0.1410
45.4	0.1355
69.8	0.1305
89.5	0.1260

accuracy is described elsewhere [8]. As for the effect of radiative heat transport in the present measurements, we have not applied any corrections to our results, because for the liquid samples studied here, we have not observed any curvature of the ΔT versus $\ln t$ lines and we could not obtain reliable optical properties, which are needed for the correction.

The present thermal conductivity data for each *n*-paraffin hydrocarbon have been correlated by means of the following equation.

$$\lambda = A' - B'T \tag{1}$$

where T represents the temperature (°C) and λ is the thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$). The coefficients which give the best representation of the present results are shown in Table VI. The table also includes the coefficients for other *n*-paraffin hydrocarbons taken from Refs. 2, 3, and 9, which are believed to be reliable measurements. The measurements for C_4, C_5, C_8, and C_{10} were taken under their saturation vapor pressures, while the others were at atmospheric pressure. However, the coefficients may not

Table IV. Thermal Conductivity of *n*-Pentadecane (n-$C_{15}H_{32}$) at Atmospheric Pressure

Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
12.9	0.1447
38.0	0.1398
62.9	0.1340
89.4	0.1290

Table V. Thermal Conductivity of *n*-Hexadecane ($n\text{-}C_{16}H_{34}$) at Atmospheric Pressure

Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
22.6	0.1440
43.8	0.1404
66.1	0.1359
89.1	0.1320

be affected by this pressure difference, since the pressure effect on the thermal conductivity $1/\lambda(\partial\lambda/\partial p)_T$ of *n*-paraffin hydrocarbons is of the order of $10^{-2}\%$ per bar [5]. As can be recognized from Table VI, both cofficients show a systematic trend with increasing carbon numbers. Figures 1 and 2 present variations of A' and B' in Eq. (1) as a function of the number of carbon atoms. In these figures, filled symbols represent the present measurements.

If we regard only the substances which have similar molecular structures, the thermophysical properties of different substances may simply be correlated in terms of parameters such as density and molecular weight. In the case of *n*-paraffin hydrocarbons, we may choose the number of carbon atoms as the parameter [10]. This parameter provides a simple and practical correlation not only for pure *n*-paraffin hydrocarbons but for their mixtures, which is shown later. Consequently, both coefficient A' and coef-

Table VI. Coefficients for the Correlation of the Thermal Conductivity of *n*-Paraffin Hydrocarbons

n-Paraffin hydrocarbon	A'	B' $\times 10^4$	Mean dev. (%)	Ref. No.
$n\text{-}C_4H_{10}$	0.1176	4.83	0.3	9
$n\text{-}C_5H_{12}$	0.1213	3.99	0.3	9
$n\text{-}C_7H_{16}$	0.1287	2.86	0.2	2
$n\text{-}C_8H_{18}$	0.1329	2.77	0.2	9
$n\text{-}C_{10}H_{22}$	0.1370	2.40	0.1	3
$n\text{-}C_{11}H_{24}$	0.1388	2.23	0.5	—
$n\text{-}C_{14}H_{30}$	0.1456	2.19	0.2	—
$n\text{-}C_{15}H_{32}$	0.1472	2.05	0.3	—
$n\text{-}C_{16}H_{34}$	0.1481	1.82	0.3	—

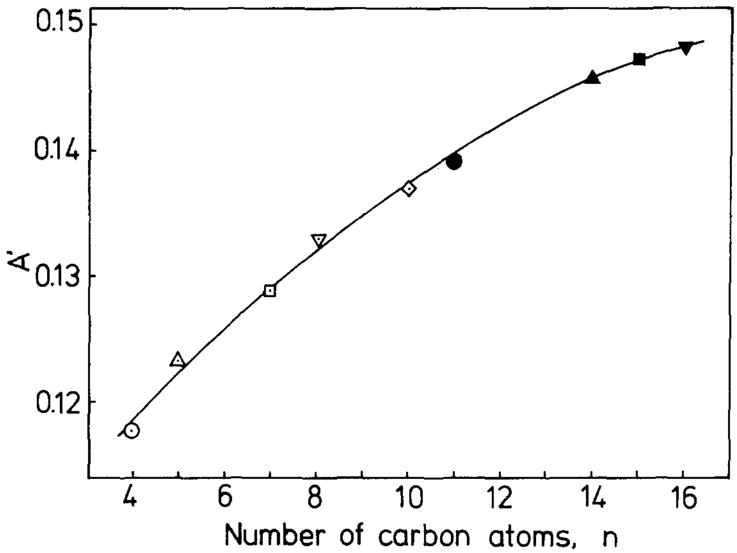

Fig. 1. Coefficient A' in Eq. (1) as a function of the number of carbon atoms. (⊙) Ref. 9; (△) Ref. 9; (⊡) Ref. 2; (▽) Ref. 9; (◇) Ref. 3; (●, ▲, ■, and ▼) present work.

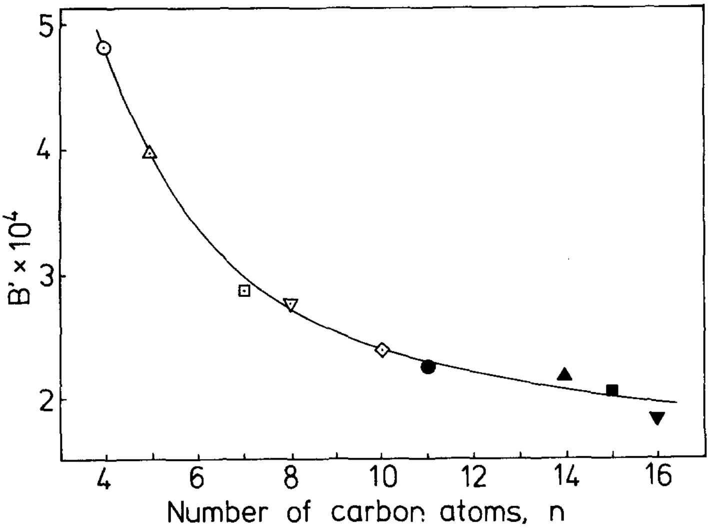

Fig. 2. Coefficient B' in Eq. (1) as a function of the number of carbon atoms. (⊙) Ref. 9; (△) Ref. 9; (⊡) Ref. 2; (▽) Ref. 9; (◇) Ref. 3; (●, ▲, ■, and ▼) present work.

ficient B' for each n-paraffin hydrocarbon are represented as a function of the number of carbon atoms n as follows.

$$\lambda = An^2 + Bn + C - [D(1/n)^2 + E(1/n) + F]T \qquad (2)$$

$A = -1.189 \times 10^{-4}$, $B = 4.860 \times 10^{-3}$, $C = 1.009 \times 10^{-1}$, $D = 2.974 \times 10^{-3}$, $E = 6.124 \times 10^{-4}$, and $F = 1.469 \times 10^{-4}$.

This equation was correlated on the basis of the experimental results for the number of carbon atoms $n = 4$, 5, 7, 8, 10, 11, 14, 15, and 16, which are listed in Table VI. Figure 3 contains plots of the deviations of the experimental data from Eq. (2) for the nine n-paraffin hydrocarbons. The mean deviation is 0.5% and the mximum deviation occurs in the case of n-C_5H_{12} at the lowest temperature but never exceeds -2.2%. Considering the fact that the individual experimental datum is accurate to within ± 1%, Eq. (2) may represent the thermal conductivity of liquid n-paraffin hydrocarbons in the temperature range of their normal liquid states within the estimated accuracy of ± 1% for the number of carbon atoms $4 \leqslant n \leqslant 16$. It must be noted that even though this correlation cannot be applied to substances other than normal paraffin hydrocarbons, it is very simple to calculate the thermal conductivity of the series of substances with an accuracy which has never been attained before.

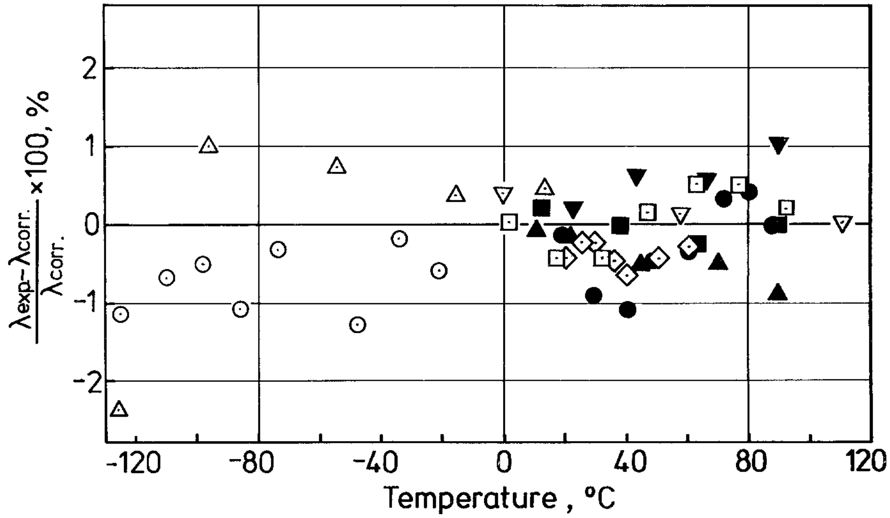

Fig. 3. Deviations of the thermal conductivity data from Eq. (2) for nine n-paraffin hydrocarbons. (⊙) C_4 [9]; (△) C_5 [9]; (□) C_7 [2]; (▽) C_8 [9]; (◇) C_{10} [3]; (●) C_{11} (present work); (▲) C_{14} (present work); (■) C_{15} (present work); (▼) C_{16} (present work).

In Fig. 4 the present correlation is compared with rather new experimental results and with other previous data on which we had not based our correlation. Recent measurements by Wakeham and his coworkers for n-C_6H_{14} [4], n-C_7H_{16} [5], n-C_8H_{18} [4], n-C_9H_{20} [6], and n-$C_{11}H_{24}$ [6], obtained by the absolute transient hot-wire method, are accurate to ± 0.3 to $\pm 0.7\%$. However, since their measurements are available only at high pressures, they were extrapolated to atmospheric pressure for comparison. As can be seen from Fig. 4, their data agree with the present correlation within the combined uncertainties. Calado et al. [7] measured the thermal conductivity of four n-paraffin hydrocarbons, C_8, C_9, C_{11}, and C_{13}, at their saturation vapor pressures with an apparatus similar to that of Wakeham et al. Due to the lack of space in Fig. 4, only mean deviations of their results from the correlation for each substance are presented: C_8, 2.3%; C_9, 1.5%; C_{11}, 2.0%; and C_{13}, 0.8%. These deviations may be understandable except for the case of n-C_8H_{18}. Kashiwagi et al. [11] measured the thermal conductivity of five hydrocarbons (C_6, C_7, C_8, C_{10}, and C_{12}) in the temperature range from 25 to 100°C using the transient hot-wire method operated in the relative mode. Their claimed accuracy is $\pm 2\%$. Their results generally agree with the present correlation, however, the deviations reach -6% in the case of C_{10} and C_{12} in the higher temperature range. The reason for this might be that they employed an X–Y recorder for the reltive measurement and the time

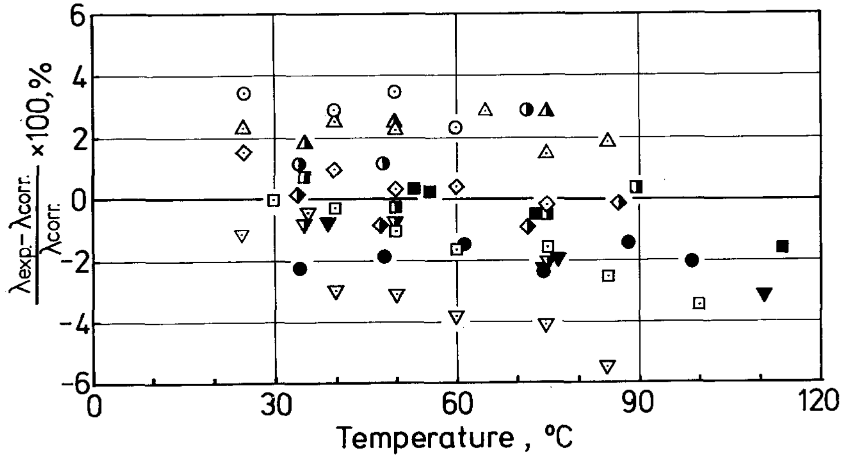

Fig. 4. Comparisons of the present correlation with other thermal conductivity data for n-paraffin hydrocarbons. (⊙) C_6 [11]; (◑) C_6 [4]; (△) C_7 [11]; (◭) C_7 [5]; (◇) C_8 [11]; (⬗) C_8 [4]; (◨) C_9 [6]; (⊡) C_{10} [11]; (⧩) C_{11} [6]; (▽) C_{12} [1]; (●) C_{13} [12]; (▼) C_{17} [13]; (■) C_{18} [13].

range for a run was considerably longer (10 s). Therefore, it may be possible that their results contain some systematic error which is larger than their claimed accuracy.

In order to see how the agreement is in the case of the number of carbon atoms not based on the present correlation, experimental results of Mustafaev [12] (C_{13}; estimated accuracy, ±5%) and by Rastorguev and Bogatov [13] (C_{17} and C_{18}; estimated accuracy, ±5%) are also plotted in Fig. 4. Even though in the latter case the number of carbon atoms is out of the range of the correlation, the mutual agreement seems to be quite reasonable.

4. RESULTS AND MIXING RULE FOR *n*-PARAFFIN HYDROCARBON MIXTURES

For the purpose of engineering applications, mixtures are being used more often than pure substances. There are almost infinite numbers of mixtures in terms of component substances and their concentrations. Consequently, in order to estimate the thermal conductivity of liquid mixtures accurately, a mixing rule, which is confirmed by systematically performd precise measurements, has to be developed. There are numerous studies on correlation techniques for the thermal conductivity of organic liquid mixtures. Some of them are summarized in Ref. 1. Most of these correlations or mixing rules employ the weight fraction as a parameter for concentration, and the departure from a linear combination rule is modified by adding some empirically determined parameters. Although these mixing rules might be applicable to a wide variety of organic liquids, their reliabilities have not been tested against a set of accurate experimental results. It may be noted that these mixing rules are not suitable for accurate estimations of the thermal conductivity of liquid mixtures for some particular groups of substances.

We still have no theoretical background on the functional form of the mixing rule for the liquid thermal conductivity, thus any form of a mixing rule can be practically acceptable as far as it is not inconsistent with experimental results. As shown in the previous section, the thermal conductivity of *n*-paraffin hydrocarbons can successfully be represented by the number of carbon atoms. We therefore have attempted to construct the following rather heuristic mixing rule for the thermal conductivity of liquid mixtures of *n*-paraffin hydrocarbons.

$$n = \sum w_i n_i \tag{3}$$

Here, w_i is the weight fraction of the ith component, whereas n_i is the number of carbon atoms for the ith component of n-paraffin hydrocarbon. Equation (3) is based on the concept that the linear mixing rule may work not directly on the thermal conductivity but indirectly on the number of carbon atoms. In other words, we assumed that the thermal conductivity of mixtures of normal hydrocarbons can be calculated by employing an equivalent number of carbon atoms determined by Eq. (3) together with Eq. (2). To verify the assumed mixing rule, we have measured the thermal conductivity of some binary and ternary mixtures of n-paraffin hydrocarbons.

As it is not possible or necessary to measure the thermal conductivity of n-paraffin hydrocarbon mixtures for all possible combinations, we selected some of the mixtures of $n\text{-}C_7H_{16}$, $n\text{-}C_{11}H_{24}$, and $n\text{-}C_{16}H_{34}$ which are in the liquid state from room temperature to about 70°C at atmospheric pressure. For binary mixtures, three kinds of combinations, namely, $n\text{-}C_7H_{16}+n\text{-}C_{11}H_{24}$, $n\text{-}C_{11}H_{24}+n\text{-}C_{16}H_{34}$, and $n\text{-}C_7H_{16}+n\text{-}C_{16}H_{34}$, were prepared with the weight fractions of 0.250, 0.500, and 0.750, respectively. In the case of ternary mixtures, only one system, $n\text{-}C_7H_{16}+n\text{-}C_{11}H_{24}+n$-

Table VII. Thermal Conductivity of $n\text{-}C_7H_{16}+n\text{-}C_{11}H_{24}$ Mixtures at Atmospheric Pressure

Weight fraction of $n\text{-}C_{11}H_{24}$	Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
0.250	23.1	0.1255
	30.7	0.1231
	40.4	0.1211
	50.2	0.1195
	60.9	0.1169
	72.3	0.1143
0.500	14.8	0.1304
	31.4	0.1260
	40.6	0.1245
	50.5	0.1214
	60.3	0.1191
	70.5	0.1160
0.750	15.4	0.1331
	31.8	0.1288
	40.6	0.1268
	50.5	0.1254
	60.6	0.1220
	70.2	0.1206

$C_{16}H_{34}$, was measured for three weight fractions, namely, 0.599:0.300:0.101, 0.100:0.299:0.601, and 0.334:0.333:0.333, respectively.

Tables VII to IX contain the measured thermal conductivity of the three binary mixtures as a function of temperature at atmospheric pressure. And the results for the ternary mixtures are listed in Table X in a similar manner. These tabulated values were obtained by averaging the raw data of 2 to 12 points under almost the same conditions. Their reproducibilities were always better than ±1 % and the accuracy of the thermal conductivity was estimated to be ±1.5 %. Although the precision of weighing for the preparation of mixtures reaches better than ±0.01 %, the accuracy of the weight fraction may be estimated to ±1 % for the binary mixtures and to ±0.5 % for the ternary mixtures. That is due to the fact that in some cases, we had to prepare the same mixtures repeatedly to the nominal values.

As for the binary mixtures, Fig. 5 shows the deviations of the present results (averaged values) from the mixing rule of Eq. (3) adopting Eq. (2). In this figure the weight fraction w represents that for the heavier component. The mean deviation is 0.7 %, whereas the maximum deviation is −1.8 %. The deviations are almost within the estimated accuracy of the

Table VIII. Thermal Conductivity of n-$C_{11}H_{24}$ + n-$C_{16}H_{34}$ Mixtures at Atmospheric Pressure

Weight fraction of n-$C_{16}H_{34}$	Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
0.250	21.4	0.1367
	31.3	0.1344
	40.9	0.1326
	50.7	0.1313
	60.7	0.1300
	70.2	0.1274
0.50	22.8	0.1390
	31.2	0.1374
	40.5	0.1369
	50.3	0.1347
	60.9	0.1325
	70.3	0.1304
0.750	24.2	0.1423
	30.0	0.1411
	40.8	0.1390
	50.3	0.1374
	60.3	0.1350
	67.5	0.1330

present measurements, ±1.5%. Figure 5 also depicts the calculated value using the linear combination rule for thermal conductivity, Eq. (4), in order to demonstrate the advantage of the present mixing rule.

$$\lambda = \sum w_i \lambda_i \tag{4}$$

For this calculation the thermal conductivities of pure components were obtained using Eq. (2), and only one example is shown in Fig. 5 for the sake of brevity. It can be seen from this figure that the mixing rule given by Eq. (4) shows a systematic departure from the experimental data reaching −2.6%, since the present results exhibit a weak convex concentration dependence rather than a linear dependence. This is the same observation as made by Ogiwara et al. [14]. Although their absolute values are considerably higher than the present correlation because of their relative measurements, the concentration dependence agrees well. We therefore may note that the proposed mixing rule, Eq. (3), offers a meaningful advantage over Eq. (4) in the case of binary mixtures.

Table IX. Thermal Conductivity of n-C_7H_{16} + n-$C_{16}H_{34}$ Mixtures at Atmospheric Pressure

Weight fraction of n-$C_{16}H_{34}$	Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
0.250	24.9	0.1271
	32.1	0.1258
	41.2	0.1240
	50.8	0.1214
	61.0	0.1192
	70.4	0.1174
0.500	16.4	0.1360
	31.5	0.1339
	40.4	0.1307
	50.7	0.1288
	61.0	0.1257
	71.3	0.1234
0.750	21.1	0.1405
	31.5	0.1377
	40.7	0.1361
	50.1	0.1341
	60.3	0.1320
	70.1	0.1289

For ternary mixtures, the deviations from the correlation of the raw data and of the calculated values by both mixing rules are shown in Fig. 6 in the case of a $C_7:C_{11}:C_{16} = 0.599:0.300:0.101$ mixture. As seen in this figure, both mixing rules exhibit almost the same applicabilities to ternary mixtures. Similar tendencies can be observed for two other mixtures. Even though the present mixing rule always calculates the thermal conductivity of the ternary mixtures systematically higher than the experimental results, the deviations never exceed +1.5%, which is consistent with the accuracy of the present data. We have not found any previous experimental studies for the ternary mixtures, thus no comparison was possible other than our own.

As a result, we may conclude that the mixing rule, Eq.(3), together with Eq. (2) can estimate the thermal conductivity of liquid mixtures of *n*-paraffin hydrocarbons within the accuracy of ±1.5%. The ranges of applicability of the present correlation for the mixtures may be from 7 to 16 for the number of carbon atoms up to 3 for the number of components. Nevertheless, the measurements have not been performed for all kinds of combinations of mixtures; the correlation might be regarded as experimen-

Table X. Thermal Conductivity of $n\text{-}C_7H_{16} + n\text{-}C_{11}H_{24} + n\text{-}C_{16}H_{34}$ Mixtures at Atmospheric Pressure

Weight fraction of $C_7:C_{11}:C_{16}$	Temperature T (°C)	Thermal conductivity λ ($W \cdot m^{-1} \cdot K^{-1}$)
0.599, 0.300, 0.101	20.6	0.1283
	31.1	0.1260
	41.2	0.1238
	50.8	0.1216
	61.5	0.1192
	70.9	0.1170
0.100, 0.299, 0.601	23.8	0.1390
	32.9	0.1371
	42.0	0.1352
	52.1	0.1331
	60.2	0.1311
	70.8	0.1285
0.334, 0.333, 0.333	20.7	0.1342
	30.3	0.1317
	40.2	0.1302
	51.0	0.1275
	59.2	0.1258
	69.6	0.1237

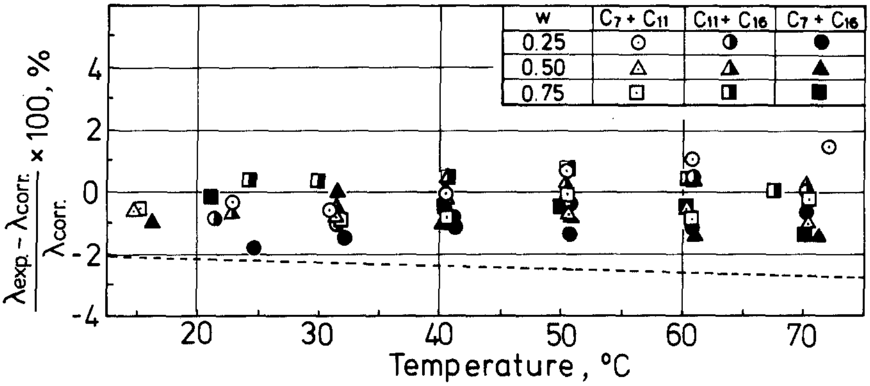

Fig. 5. Deviations of the present thermal conductivity data from Eq. (3) together with Eq. (2) for binary mixtures of *n*-paraffin hydrocarbons. (- - -) Calculated from Eq. (4) in the case of $C_{11}+C_{16}$ ($w_{C_{11}}=0.500$) mixture.

tally verified. This is because we have limited the application of the correlation to a series of similar substances, namely, *n*-paraffin hydrocarbons. We have not attempted to measure the thermal conductivity of mixtures of more than four components, however, considering that no anomalous phenomena can be expected as in the case of multicomponent systems just as for mixtures of up to three components, the present mixing rule might be useful for multicomponent mixtures.

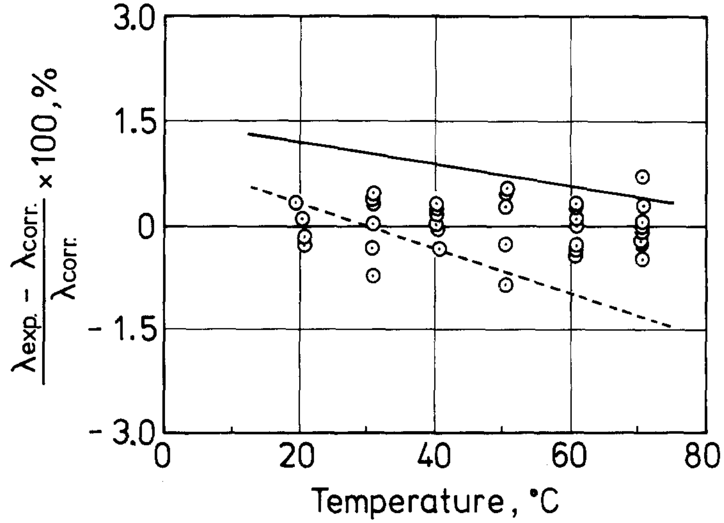

Fig. 6. Comparisons of the mixing rules with the present thermal conductivity data for ternary mixture of *n*-paraffin hydrocarbons (C_7:C_{11}:C_{16} = 0.599:0.300:0.101). (——) Eq. (3); (– – –) Eq. (4).

5. CONCLUSIONS

Based on systematically operated measurements, a new generalized correlation and a mixing rule for the thermal conductivity of liquid *n*-paraffin hydrocarbons and their mixtures have been proposed. The correlation for pure *n*-paraffins, Eq. (2), can be used in the temperature range of their normal liquid states with the number of carbon atoms $4 \leqslant n \leqslant 16$. The accuracy is estimated to be $\pm 1\%$. For mixtures the mixing rule, Eq. (3), with Eq. (2) was verified to estimate their thermal conductivity within the accuracy of $\pm 1.5\%$ in the range of the number of carbon atoms $7 \leqslant n \leqslant 16$ up to ternary mixtures. This range of validity might be extrapolated with regard to the number of carbon atoms and the number of components as long as we are dealing with normal paraffin hydrocarbons. For branched hydrocarbons, however, further experimental studies will be needed.

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