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Thermal Conductivity of Liquid *n*-Alkanes

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The thermal conductivity of liquids has been shown in the past to be difficult to predict with a reasonable accuracy, due to the lack of accurate experimental data and reliable prediction schemes. However, data of a high accuracy, and covering wide density ranges, obtained recently in laboratories in Boulder, Lisbon, and London with the transient hot-wire technique, can be used to revise an existing correlation scheme and to develop a new universal predictive technique for the thermal conductivity of liquid normal alkanes. The proposed correlation scheme is constructed on a theoretically based treatment of the van der Waals model of a liquid, which permits the prediction of the density dependence and the thermal conductivity of liquid *n*-alkanes, methane to tridecane, for temperatures between 110 and 370 K and pressures up to 0.6 MPa, i.e., for $0.3 \leqslant T/T_c \leqslant 0.7$ and $2.4 \leqslant \rho/\rho_c \leqslant 3.7$, with an accuracy of $\pm 1\,\%$, given a known value of the thermal conductivity of the fluid at the desired temperature. A generalization of the hard-core volumes obtained, as a function of the number of carbon atoms, showed that it was possible to predict the thermal conductivity of pentane to tetradecane $\pm 2\,\%$, without the necessity of available experimental measurements.

KEY WORDS: correlation; *n*-alkanes; thermal conductivity.

1. INTRODUCTION

The transport properties of liquids have been very difficult to be understood on any significant molecular basis, especially thermal conductivity.

Recently, an increased demand for accurate thermophysical property data has been observed as a result of the industrial needs for a better design of individual items of a chemical plant, as well as for energy

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management optimization of chemical processes [1]. Moreover, the effect of inaccurate data of thermophysical properties of fluids on the design and operation of different units of chemical processing has been studied by several authors [2–12] and emphasized the need for accurate experimental data for optimal technological design and efficient plant operation.

It is evidently impossible for experimental measurements of these properties ever to be carried out over the entire range of thermodynamic states for every pure component and for multicomponent mixtures of arbitrary composition. Therefore, it is necessary to have some means available whereby the existing experimental data may be employed to generate the required quantities.

The measurement of the thermal conductivity of fluids has developed considerably in the last decade, owing to the establishment of a highly accurate measurement technique, the transient hot-wire method [13]. Data obtained mainly with this technique have been recently proposed as standard reference data for the thermal conductivity of liquids [14]. Highly accurate data for the thermal conductivity of *n*-alkanes, from methane to tridecane, have been recently obtained using this technique and, therefore, used to establish a reliable prediction scheme, with some theoretical foundation.

In the last two decades there has been considerable progress towards molecular description of the transport properties of fluids [15–17], one significant factor in this development being the computer simulation studies of transport processes by molecular dynamics [18, 42]. In particular, the van der Waals model of a fluid [15] was a natural extension of the work of Alder and Wainwright [18]. In this model the intermolecular pair potential consists of a rigid core surrounded by a nearly uniform, long-range, attractive potential. The molecular motion can thus be approximated by a succession of linear trajectories and hard-core collisions.

The application of this model to real systems requires a temperature-dependent core size volume, V_0. The use of the rigid-sphere model allows the dominant effects, in transport phenomena in dense fluids, to be analyzed with the aid of the Enskog theory, corrected for correlations between velocities of the molecules from molecular dynamics simulations. Strictly speaking, this model is applicable only to monatomic systems. However, in the next section, it is shown how an heuristic extension of the van der Waals model, previously suggested by Menashe et al. [19], can be applied to polyatomic liquids and to the prediction of the thermal conductivity of *n*-alkanes over wide temperature and pressure ranges.

Although we use the word *prediction* here to designate the proposed schemes of property evaluation, care must be taken in its use as such, even though it is the word most currently used in chemical engineering. The joint use of theory and experiment recommends the designation of semi-empirical estimation for such a procedure. For a complete discussion of this problem, please see Ref. 43.

2. THE van der Waals MODEL

The van der Waals model for transport properties of fluids incorporates a hard-sphere theory for dense fluids, with a temperature-dependent core size, V_0 [15–17]. Its basis and application to liquids have been tried with some success, and details of it can be found in Ref. 17. It has been shown recently that the thermal conductivity of liquid argon and liquid methane can be correlated by this model [20, 23] to within $\pm 2\%$.

The main result of the model consists in defining a reduced thermal conductivity λ^* by

$$\lambda^* = \frac{\lambda}{\lambda_0}\left(\frac{V}{V_0}\right)^{2/3} = F_\lambda\left(\frac{V}{V_0}\right) \tag{1}$$

where λ_0 is the hard-sphere thermal conductivity at temperature T and V is the molar volume of the fluid. This reduced thermal conductivity is a function of the ratio V/V_0 alone, given for monatomic liquids by [15, 20]

$$F_\lambda\left(\frac{V}{V_0}\right) = [1.02 + 0.1(x - 0.3)]\left\{\frac{(1-0.7405x)^3}{(1-0.3702x)} + 3.554x + 6.624x^2\left[\frac{(1-0.3702x)}{(1-0.7405x)^3}\right]\right\}x^{-2/3} \tag{2}$$

where $x = V/V_0$.

If the hard-sphere model represents the thermal conductivity of any liquid alkanes, as has already been shown by Menashe et al. [19], we can obtain a reduced λ^* from experimental data with the help of Eq. (1). We obtain

$$\lambda^* = 1.610 \cdot 10^8 \lambda V^{2/3}\left(\frac{M}{R^3 T}\right)^{1/2} \tag{3}$$

where M is the molecular mass, R is the gas constant, and all quantities are expressed in SI units.

The application of Eqs. (1) to (3) to polyatomic fluids requires a fundamental modification introduced by Menashe et al. [19]. Polyatomic fluids have internal energy in addition to translational energy. The contribution of the internal degrees of freedom to the thermal conductivity was assumed to have the same density dependence as the translational part. Therefore the quantity

$$\lambda' = \frac{\lambda}{\lambda_0}\left(\frac{V}{V_0}\right)^{2/3} \tag{4}$$

has exactly the same dependence on V/V_0 as does λ^* for monatomic fluids, but λ_0 is now the zero-density thermal conductivity of the fluid, evaluated by

$$\lambda_0 = \frac{15}{4}\frac{R}{M}\left(1 + 0.352\,\frac{C^0_{v,\mathrm{int}}}{R}\right)\eta_0 \tag{5}$$

where $C^0_{v,\mathrm{int}}$ is the internal part of the molar heat capacity at zero density and η_0 is the hard-sphere viscosity. With this reformulation λ' is given by

$$\lambda' = \frac{1.610 \cdot 10^8 \lambda V^{2/3} (M/R^3 T)^{1/2}}{1 + 0.352\,(C^0_{v,\mathrm{int}}/R)} \tag{6}$$

and it contains only experimentally accessible quantities. Thus, according to the preceding hypothesis, it should be possible to superimpose plots of experimental curves of λ' against $\ln V$ for a single fluid at a series of temperatures merely by imposing relative shifts along the $\ln V$ axis.

Menashe et al. [19] tested the fundamental hypothesis of the model with experimental data on propane, *n*-heptane, *n*-nonane, *n*-undecane, and *n*-tridecane and concluded that it was possible to superimpose the experimental curves of λ' as a function of $\ln V$ with the theoretical curves. They thus obtained V_0 for a given temperature. Furthermore, they found that the same procedure was possible for several alkanes to within $\pm 1\,\%$ of λ and a standard deviation of 0.35% and proposed a universal relation for λ' as a function of V/V_0.

After this work was published several new sets of experimental data have been obtained by different laboratories that permit a more stringent test of the heuristic extension of the van der Waals model. Furthermore, at the time, values of the thermal conductivity of the *n*-alkanes used had a correction from radiative heat transport in the cells, later proved to be unnecessary [21].

We present in the next section a revised correlation, as well as a generalized equation which can be very useful for predictive purposes.

3. THE CORRELATIONS

In the next two sections we present two different schemes for the prediction of the thermal conductivity of the *n*-alkanes.

Table I. Values of Hard-Core Volumes Obtained for the Different *n*-Alkanes at Several Temperatures

n-Alkane	T (K)	$10^6 V_0$ ($\mathrm{m^3 \cdot mol^{-1}}$)	T_r
Methane [22]	110	30.7	0.58
	135	30.1	0.71
	155	29.8	0.81
	175	30.1	0.92
Methane [23]	110	30.7	0.58
	125	30.3	0.66
	155	30.4	0.81
	180	32.9	0.94
Ethane [24]	110	51.6	0.36
	135	49.4	0.44
	155	47.6	0.51
	175	46.0	0.57
	195	44.5	0.64
	215	43.1	0.70
	235	42.0	0.77
	255	41.3	0.83
	275	40.5	0.90
	295	40.6	0.97
Propane [25]	111	68.6	0.30
	139	64.5	0.38
	169	60.3	0.46
	196	57.1	0.53
	227	53.7	0.61
	260	51.3	0.70
	289	48.4	0.81
Butane [26]	258.2	58.0	0.70
	358.2	54.8	0.84
	383.2	56.3	0.90
	413.2	59.2	0.97
Pentane [27]	305.8	67.4	0.65
	322.8	65.9	0.69
	342.5	64.2	0.73
Hexane [28]	307.2	78.7	0.60
	321.2	77.3	0.63
	345.2	74.9	0.68
	360.2	73.4	0.71
Heptane [29]	308.2	89.3	0.57
	323.2	87.4	0.60
	348.2	84.4	0.64
Octane [28]	307.2	100.7	0.54
	321.2	98.6	0.56
	345.2	95.3	0.61
	362.2	93.1	0.64
Nonane [30]	308.2	112.0	0.52
	323.2	109.6	0.54
	348.2	106.0	0.58
	363.2	104.0	0.61
Undecane [30]	308.2	135.5	0.48
	323.2	132.6	0.50
	348.2	128.1	0.54
Tridecane [31]	308.2	159.4	0.45
	321.2	155.9	0.47
	346.2	150.4	0.51

3.1. Scheme I

Using the superposition method described in the previous section, we have obtained $V_0(T)$ values for normal alkanes from C1 to C9, C11, and C13, using recently published data [22–31]. These data cover a wide range of states, from 100 to 300 K and pressures up to 70 MPa [22–25], from 300 to 500 K and pressures up to 70 MPa [26], and from 300 to 380 K with pressures up to 500 MPa [27–31]. All these data are accurate to within ±1% or less. The values of the hard-core volumes obtained are presented in Table I, together with the reduced temperature for each isotherm; for all the *n*-alkanes studied.

The data used for the densities of each hydrocarbon can be found in the original papers with thermal conductivity data (see Table I). The densities of these fluids can also be predicted at temperatures up to $0.66T_c$ and pressures up to 150 MPa using the Tait equation, with parameters developed by Dymond and Malhotra [32] to within 0.2%.

The heat capacities at zero density were taken from several sources, namely, methane [33], ethane [34], propane [35], and *n*-butane to tridecane [36]. The data $T_r \leqslant 0.7$, where $T_r = T/T_c$ is the reduced temperature, can be described by the equation

$$\ln \lambda' = 2.87337 - 2.2709 \ln\left(\frac{V}{V_0}\right) \tag{7}$$

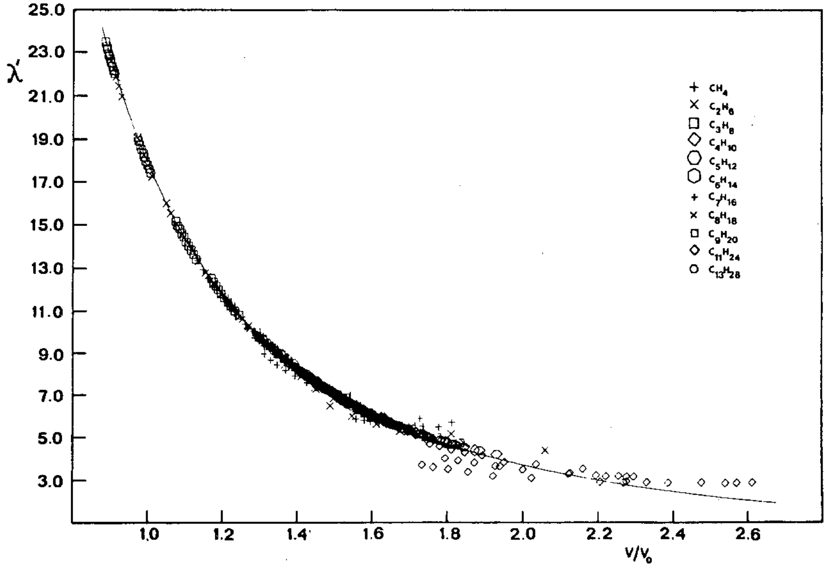

Fig. 1. Plot of the reduced thermal conductivity λ' as a function of V/V_0. The curve is represented by Eq. (7).

with a standard deviation of 0.4% and a maximum deviation of 2.2%. Figure 1 represents all the data of λ' as a function of V/V_0 together with Eq. (7), and Fig. 2 shows a deviation plot for the same data but only for $T_r \leqslant 0.7$.

For the high-temperature isotherms ($T_r > 0.7$), the deviations are much higher, increasing with the reduced temperature. The highest deviation was found for the *n*-butane isotherm at $T_r = 0.97$ (up to $\pm 30\%$). The results already found for correlations involving other thermophysical properties show that the shape of the molecules cannot be neglected for $T_r > 0.7$. This seems to be confirmed by the present study. This behavior is presumably an immediate consequence of the fact that these regions are specifically excluded from the model. The fact already obsrved for different fluids that the excess thermal conductivity of liquids is a function of temperature for high densities and high temperatures cannot be taken into account by the van der Waals model [44] and is probably derived from the increasing contribution of the attractive forces for liquid-phase heat transport, as the critical point is approached. A full discussion of this behavior can be found in Ref. 39.

Using Eq. (7) and the values of $V_0(T)$ included in Table I, the value of the thermal conductivity for any of these hydrocarbons in the liquid state can be estimated to within 0.8% at a 2σ level (σ is the standard deviation), if the density is known, for the temperatures presented. For any other temperature, a value of the thermal conductivity at any density is also needed to obtain from Eq. (7) the corresponding value of V_0. As an alternative, linear fits of $V_0(T)$ in Table I can be made (except for propane, where a quadratic fit is needed) for the temperature range referred. Such a procedure lowers the accuracy of the prediction to about 1.5%.

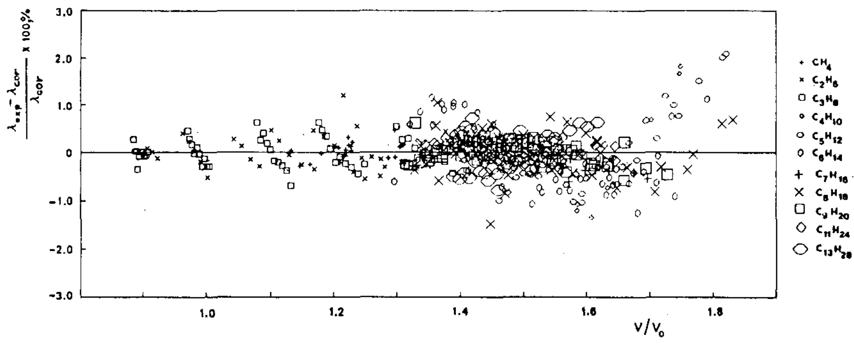

Fig. 2. Deviation plot of experimental data from Eq. (7) for $T_r \leqslant 0.7$ (scheme I).

3.2. Scheme II

The prediction scheme presented so far is a very powerful method for the interpolation of data for the *n*-alkanes studied, for the interval $0.3 \leqslant T_r \leqslant 0.7$ and $2.4 \leqslant \rho_r \leqslant 3.7$, where ρ_r is the reduced density ρ/ρ_c. However, it is limited by the fact that one requires a pair of values $(\lambda_{exp}, \rho_{exp})$ to use the scheme for any temperature other than the reported ones.

In many engineering applications this is not the case, and it will be very convenient to predict the thermal conductivity of other *n*-alkanes not used in the correlation derivation and for which there are not extensive and accurate data.

Figure 3 shows the variation of the hard-core volumes V_0 with temperature for the hydrocarbons that are liquid around room temperature

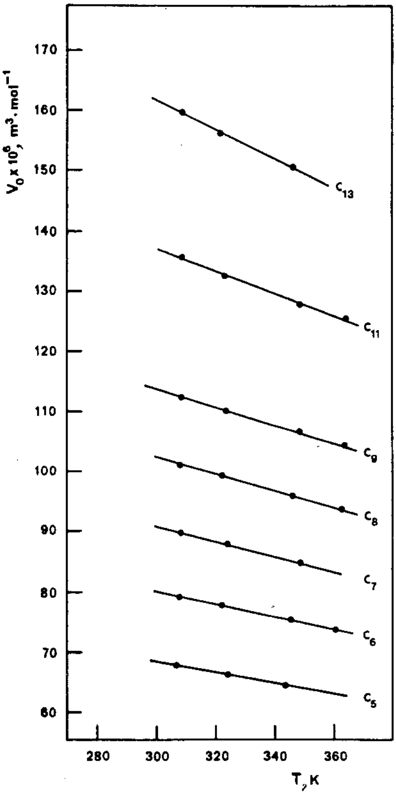

Fig. 3. Temperature dependence of hard-core volumes for *n*-alkanes ($n_c \geqslant 5$). Lines are given by Eq. (8).

and normal pressures, i.e., $n_c \geqslant 5$, where n_c is the number of carbon atoms in each *n*-alkane molecule. It is also possible to find the same type of behavior for methane to butane, but the following analysis did not apply to them within the correlation accuracy we were aiming to find, possibly due to the fact that these small molecules are more sensitive to structural effects than the big chain molecules. It can be easily seen that V_0 varies linearly with temperature and that, furthermore, the value of V_0 for a given temperature is a linear function of the number of carbon atoms, n_c. This being the case we can write

$$10^6 V_0 = v'(n_c) + v''(n_c)\,T \tag{8}$$

where v' and v'' are linear functions of the number of carbon atoms, as shown in Fig. 4, V_0 is in $\mathrm{m^3 \cdot mol^{-1}}$, and T is in K. It was found that

$$v'(n_c) = 7.39 + 17.003 n_c \tag{9}$$

$$v''(n_c) = 0.006 - 0.01788 n_c \tag{10}$$

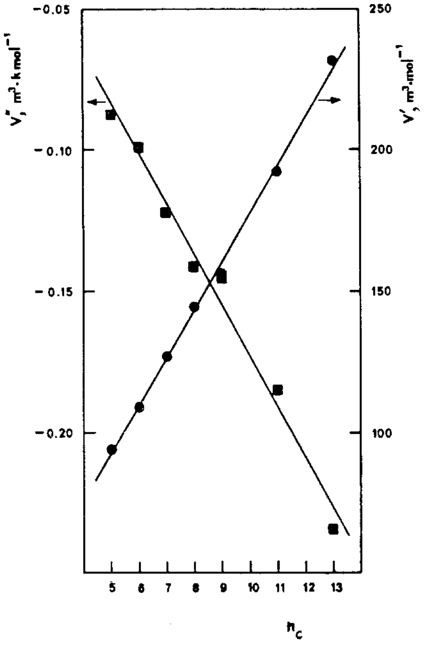

Fig. 4. The intercept and the slope of Eq. (8) as a function of the number of carbon atoms in the *n*-alkane molecule. The lines are given by Eqs. (9) and (10).

and that the values presented in Table I could predicted to within ±1% using Eqs. (8) to (10).

An equation for $\lambda'(V/V_0)$ has been optimized only for the *n*-alkanes covered by the present scheme and it was found that

$$\ln \lambda' = 2.8699 - 2.2623 \ln \left(\frac{V}{V_0}\right) \tag{11}$$

with a standard deviation of 0.45% and a maximum deviation of 2.5%, when the values of V_0 are taken from Table I. If Eqs. (8) to (10) are used to predict the values of V_0, the maximum deviation becames 4.2% and the standard deviation is of the order of 1%. We can therefore say that, within a 2σ level, the accuracy of the prediction is 2%. Figure 5 shows the overall dispersion diagram for all the data used in this second scheme. It is evident that the prediction is less accurate than that obtained with scheme I, but it can be much more useful in the absence of any experimental data. Furthermore, it can be used to predict data for other *n*-alkanes, not used to develop the prediction scheme, or for all the *n*-alkanes on the saturation line. It should be emphasized that the data to be compared in the saturation line are far away from the lowest pressure used in the establishment of equation (11).

Nieto de Castro et al. [37, 38] presented data for *n*-heptane and *n*-decane, and Fareleira et al. [39, 40] measured the thermal conductivity of *n*-octane, *n*-nonane, *n*-undecane, and *n*-tetradecane. Recently Assael et al. [41] measured *n*-hexane, *n*-heptańe, and *n*-decane. All the experimental data have an accuaracy better than 1%. Densities and C_p^0 were taken from

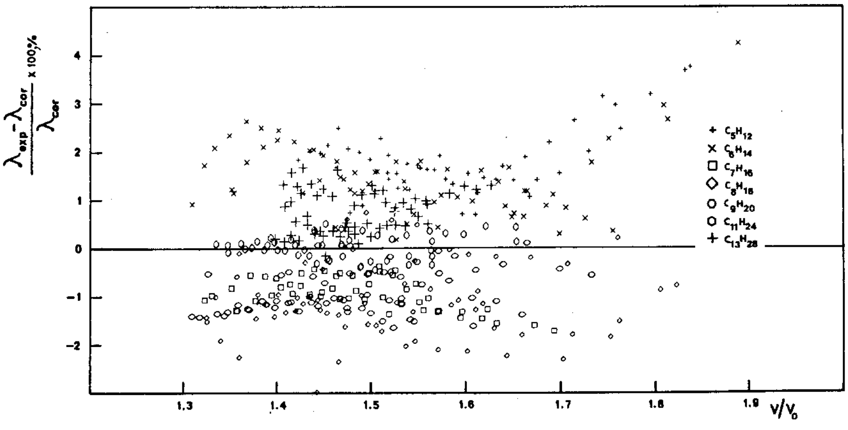

Fig. 5. Deviation plot for scheme II prediction as a function of V/V_0 for all the *n*-alkanes used in the correlation given by Eq. (11).

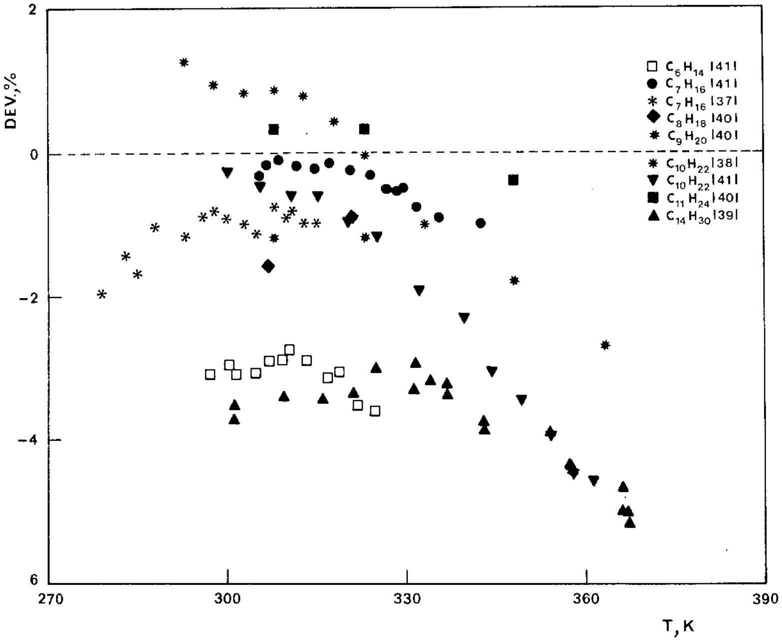

Fig. 6. Comparison between predicted data for the saturation line, using scheme II, for several *n*-alkanes. The deviation is defined as $(\lambda_{\text{corr}} - \lambda_{\text{exp}})/\lambda_{\text{exp}} \times 100$ in %.

Ref. 36. Figure 6 shows a comparison between the predicted and the experimental data. It can be seen that, despite the well-known decrease in the curvature of the line λ vs ρ as saturation is approached, the correlation is capable of predicting the experimental values with a maximum deviation of 5% for *n*-decane and *n*-tetradecane at high temperatures.

4. CONCLUSIONS

It has been shown that a heuristic extension of the van der Waals model was capable of predicting the thermal conductivity of *n*-alkanes, methane to nonane, undecane, and tridecane, within 0.8% (at a 2σ level) (scheme I) in the temperature range $0.3 \leqslant T_r \leqslant 0.7$ and density range $2.4 \leqslant \rho_r \leqslant 3.7$. This scheme needs the a priori knowledge of at least one pair of values of thermal conductivity and density for the desired temperature.

A generalization of the dependence on temperature and number of carbon atoms of the hard-core volumes allowed the development of a correlation, valid in the same temperature and pressure ranges of scheme I, based on *n*-pentane to *n*-nonane, *n*-undecane, and *n*-tridecane, that is capable of predicting the thermal conductivity of all *n*-alkanes, from pentane to tetradecane, with a 2% accuracy (at a 2σ level) and a maximum deviation of about 4% (scheme II).

The correlation was used to predict the thermal conductivity of *n*-alkanes along the saturation line and the results obtained seem to confirm the overall accuracy of the correlation.

The full predictive power of a correlation of this type can be achieved only when equations of state of *n*-alkanes are available. Unfortunately, only the recent effort of prediction of densities of *n*-alkanes as a function of temperature and pressure with a high accuracy is available [32], and only for pressures up to 150 MPa.

For most engineering calculations this range is the most useful and the use of the proposed scheme jointly with the density prediction scheme of Dymond and Malhotra [32] seems to be the most reliable predictive method for thermal conductivity presented so far, covering a wide range of temperatures and pressures.

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