

Reference Correlation of the Thermal Conductivity of *n*-Heptane from the Triple Point to 600 K and up to 250 MPa

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M. J. Assael, I. Bogdanou, S. K. Mylona, M. L. Huber, R. A. Perkins, and V. Vesovic



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Reference Correlation of the Thermal Conductivity of *n*-Heptane from the Triple Point to 600 K and up to 250 MPa

M. J. Assael^{a)} I. Bogdanou, and S. K. Mylonas

Laboratory of Thermophysical Properties and Environmental Processes, Chemical Engineering Department, Aristotle University, Thessaloniki 54124, Greece

M. L. Huber and R. A. Perkins

Applied Chemicals and Materials Division, National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305, USA

V. Vesovic

Department of Earth Science and Engineering, Imperial College London, London SW7 2AZ, United Kingdom

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This paper contains new, representative reference equations for the thermal conductivity of *n*-heptane. The equations are based in part upon a body of experimental data that have been critically assessed for internal consistency and for agreement with theory whenever possible. In the case of the dilute-gas thermal conductivity, a theoretically based correlation was adopted in order to extend the temperature range of the experimental data. Moreover, in the critical region, the experimentally observed enhancement of the thermal conductivity is well represented by theoretically based equations containing just one adjustable parameter. The correlations are applicable for the temperature range from the triple point to 600 K and pressures up to 250 MPa. The overall uncertainty (considered to be estimates of a combined expanded uncertainty with a coverage factor of 2) of the proposed correlation is estimated, for pressures less than 250 MPa and temperatures less than 600 K, to be less than 4%. © 2013 by the U.S. Secretary of Commerce on behalf of the United States. All rights reserved. [<http://dx.doi.org/10.1063/1.4794091>]

Key words: critical phenomena; *n*-heptane; thermal conductivity; transport properties.

CONTENTS

1. Introduction	2
2. Methodology.....	2
3. The Correlation.....	2
3.1. The dilute-gas limit	4
3.2. The residual thermal conductivity.....	5
3.3. The critical enhancement	6
3.3.1. Simplified crossover model	6
3.3.2. Empirical critical enhancement	8
4. Computer-Program Verification.....	8
5. Conclusion	8
Acknowledgments	8
6. References	8

List of Tables

1. Thermal conductivity measurements of <i>n</i> -heptane	3
2. Coefficients of Eq. (7) for the residual thermal conductivity of <i>n</i> -heptane	6
3. Evaluation of the <i>n</i> -heptane thermal-conductivity correlation for the primary data	6
4. Sample points for computer verification of the correlating equations	8

List of Figures

1. Temperature and pressure ranges of the primary experimental thermal conductivity data for <i>n</i> -heptane.	4
2. Dilute-gas limit thermal conductivity as a function of temperature.....	5
3. Percentage deviations of primary experimental data for dilute-gas limit thermal conductivity of <i>n</i> -heptane from the values calculated by Eqs. (3)–(5).	5

^{a)} Author to whom correspondence should be addressed; electronic mail: assael@auth.gr.
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4. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of density. 6
5. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature. . . . 7
6. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of pressure. 7
7. Percentage deviations of secondary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature (period 1969–1997). 7
8. Percentage deviations of secondary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature (period 1913–1968). 7
9. Thermal conductivity of *n*-heptane as a function of temperature for different pressures. 8
10. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by Eqs. (1), (6), and (7) and the empirical enhancement of Eq. (13) as a function of temperature. . . 8

1. Introduction

In a series of recent papers, new reference correlations for the thermal conductivity of normal and parahydrogen,¹ SF₆,² toluene,³ benzene,⁴ and *n*-hexane,⁵ covering wide ranges of conditions of temperature and pressure, were reported. In this paper, the work is extended to the thermal conductivity of *n*-heptane.

The goal of this work is to critically assess the available literature data, and provide a wide-ranging correlation for the thermal conductivity of *n*-heptane that is valid over gas, liquid, and supercritical states, and that incorporates densities provided by the equation of state (EOS) of Span and Wagner.⁶ It should further be noted that although there are some alternative approaches today for such correlations (e.g., neural networks), we followed a procedure based upon kinetic theory and our previous work, as will be discussed in Secs. 2–4.

2. Methodology

The thermal conductivity λ is expressed as the sum of three independent contributions, as

$$\lambda(\rho, T) = \lambda_o(T) + \Delta\lambda(\rho, T) + \Delta\lambda_c(\rho, T), \quad (1)$$

where ρ is the density, T is the temperature, and the first term, $\lambda_o(T) = \lambda(0, T)$, is the contribution to the thermal conductivity in the dilute-gas limit, where only two-body molecular interactions occur. The final term, $\Delta\lambda_c(\rho, T)$, the critical enhancement, arises from the long-range density fluctuations that occur in a fluid near its critical point, which contribute to a divergence of the thermal conductivity at the critical point.

Finally, the term $\Delta\lambda(\rho, T)$, the residual property, represents the contribution of all other effects to the thermal conductivity of the fluid at elevated densities including many-body collisions, molecular-velocity correlations, and collisional transfer.

The identification of these three separate contributions to the thermal conductivity and to transport properties in general is useful because it is possible, to some extent, to treat both $\lambda_o(T)$ and $\Delta\lambda_c(\rho, T)$ theoretically. In addition, it is possible to derive information about $\lambda_o(T)$ from experiment. In contrast, there is almost no theoretical guidance concerning the residual contribution, $\Delta\lambda(\rho, T)$, so that its evaluation is based entirely on experimentally obtained data.

The analysis described above should be applied to the best available experimental data for the thermal conductivity. Thus, a prerequisite to the analysis is a critical assessment of the experimental data. For this purpose, two categories of experimental data are defined: primary data employed in the development of the correlation, and secondary data used simply for comparison purposes. According to the recommendation adopted by the Subcommittee on Transport Properties (now known as The International Association for Transport Properties) of the International Union of Pure and Applied Chemistry, the primary data are identified by a well-established set of criteria.⁷ These criteria have been successfully employed to establish standard reference values for the viscosity and thermal conductivity of fluids over wide ranges of conditions, with uncertainties in the range of 1%. However, in many cases, such a narrow definition unacceptably limits the range of the data representation. Consequently, within the primary data set, it is also necessary to include results that extend over a wide range of conditions, albeit with poorer accuracy, provided they are consistent with other more accurate data or with theory. In all cases, the accuracy claimed for the final recommended data must reflect the estimated uncertainty in the primary information.

3. The Correlation

Table 1 summarizes, to the best of our knowledge, the experimental measurements^{8–50} of the thermal conductivity of *n*-heptane reported in the literature. Forty-three sets are included in the table. From these sets, 12 were considered as primary data.

The data of Assael *et al.*,¹⁰ Menashe and Wakeham,¹⁴ Nagasaka and Nagashima,¹⁵ and Nieto de Castro *et al.*¹⁷ were obtained in absolute transient hot-wire instruments with an uncertainty of less than 0.5%, and these four sets were considered as primary data. Also in the primary data set, the measurements of Tanaka *et al.*,⁹ Forsman *et al.*,¹² Kandiyoti *et al.*,¹⁸ and Mukhamedzyanov *et al.*,¹⁹ performed in absolute transient hot-wire instruments with a slightly inferior uncertainty, up to 2%, were included. Two more transient hot-wire sets were considered as primary data; the relative measurements of Watanabe and Seong⁸ and Kashiwagi *et al.*,¹³ with uncertainty of 0.5% and 2%, respectively. Finally, the latest measurements of Naziev *et al.*,^{11,16} obtained in an absolute

TABLE 1. Thermal conductivity measurements of *n*-heptane

First author	Year publ.	Technique employed ^a	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Primary data							
Watanabe ⁸	2002	THW (Rel)	98.00	0.5	24	261–337	0.101
Tanaka ⁹	1988	THW (Abs)	99.00	1	43	283–374	0.1–250
Assael ¹⁰	1987	THW (Abs)	99.50	0.5	14	305–343	0.101
Naziev ^{11,b}	1984	CC	na	na	36	425–624	0.101–50
Forsman ¹²	1982	THW (Abs)	na	2	42	269–328	66–1051
Kashiwagi ¹³	1982	THW (Rel)	99.00	2	6	298–359	0.101
Menashe ¹⁴	1981	THW (Abs)	99.50	0.5	50	308–349	52–500
Nagasaka ¹⁵	1981	THW (Abs)	99.00	0.5	7	274–366	0.101
Naziev ¹⁶	1981	CC (Abs)	na	1.5	35	200–288	0.101–50
Nieto de Castro ¹⁷	1976	THW (Abs)	99.50	0.3	11	293–304	0.004–0.008
Kandiyoti ¹⁸	1972	THW (Abs)	99.91	1.3	17	191–383	0–0.136
Mukhamedzyanov ¹⁹	1971	THW (Rel)	99.45	2	73	298–449	0.098–226
Secondary data							
Lei ²⁰	1997	THW (Abs)	99.50	na	6	253–304	0.101
Tong ²¹	1995	ThRes (Rel)	na	1	19	323–524	5–18
Rowley ²²	1988	THW	99.95	2	1	303	0.101
Knibbe ²³	1987	THW (Abs)	na	2	9	286–444	0.003–0.57
Fischer ²⁴	1985	PP	99.00	2	5	253–474	0–1
El-Sharkawy ²⁵	1983	ACHW (Abs)	98.80	1.5–2	9	293–374	0.004–0.106
Shashkov ^{26,b}	1983	HF (Abs)	99.50	1.5	5	331–374	0.003
Magerramov ²⁷	1982	SSCS	99.96	2.5	50	286–498	0.101–50
Atalla ²⁸	1981	ACHW (Abs)	na	2.2	7	283–313	0.101
Guseinov ²⁹	1980	CC (Abs)	99.80	1.4–2.7	24	296–483	0.1–50
Ogiwara ³⁰	1980	SSPP (Rel)	99.99	na	5	293–333	0.101
Tarzmanov ^{31,b}	1974	HW	na	1.5	31	367–693	0.008–0.1
Mallan ³²	1972	THW	na	2.5	6	298–364	0.101
Naziev ³³	1971	CC (Abs)	na	1.4	129	293–574	0.101–100
Kerimov ³⁴	1970	HW	na	1.5	3	299–354	0.101
Bogatov ³⁵	1969	SSHW	na	1.5	54	293–473	0.1–49
Brykov ³⁶	1968	CC (Rel)	na	0.5	19	193–360	0.101
Abas-Zade ³⁷	1967	SSHW	na	na	4	286–363	0.101
Tarzmanov ³⁸	1967	HW	na	1	12	367–688	0.101
Carmichael ^{39,b}	1966	SSCS	99.89	2	2	294–378	0.003
Jobst ⁴⁰	1964	THW (Abs)	na	2	5	182–321	0.101
Akhmedov ⁴¹	1963	Cal	na	na	2	293–324	0.101
Mukhamedzyanov ⁴²	1963	THW (Rel)	na	1	9	309–364	0.101
Golubev ^{43,b}	1962	CC	na	na	153	293–634	0.098–59
Vilim ⁴⁴	1960	CC	na	2	1	293	0.101
Frontasev ⁴⁵	1959	THW (Abs)	na	2	2	293–304	0.101
Briggs ⁴⁶	1957	CC (Rel)	na	3	5	293–334	0.101
Lambert ^{47,b}	1955	HW (Rel)	na	0.5	1	339	0.0351
Sakiadis ⁴⁸	1955	SSPP	99.00	1.5	10	305–350	0.101
Filippov ⁴⁹	1954	PP	na	2–3	5	286–364	0.101
Hernqvist ⁵⁰	1913	na	na	na	6	276–278	0.101

^aAbs, absolute; ACHW, AC hot wire; Cal, calorimetry; CC, coaxial cylinder; HF, hot filament; HW, hot wire; na, not available; PP, parallel plate; Rel, relative; SSCS, steady-state concentric spheres; SSHW, steady-state hot wire; SSPP, steady-state parallel plate; ThRes, thermal resistor; THW, transient hot wire.

^bIncludes vapor data employed to derive the dilute-gas limit thermal-conductivity correlation.

concentric-cylinder instrument, were also included in the primary data set.

Figure 1 shows the temperature and pressure range of the primary measurements outlined in Table 1. Temperatures for all data were converted to the ITS-90 temperature scale.⁵¹ The development of the correlation requires densities; Span and Wagner⁶ in 2003 reviewed the thermodynamic properties of *n*-heptane and developed an accurate, wide-ranging EOS from the triple point up to 600 K and 100 MPa, with an uncertainty of 0.2% in density. Recently, Sagdeev *et al.*⁵² measured the

density of *n*-heptane at 298 and 470 K and up to 250 MPa, with an uncertainty of better than 0.3% at high pressures. Sagdeev *et al.*⁵² reported that their measurements agreed with the Span and Wagner⁶ densities up to 250 MPa, within 0.07%–0.25%. Hence, the Span and Wagner⁶ equation was employed up to 600 K and 250 MPa—limits that hence also restrict the present thermal conductivity correlation. Note that the experimental data in Table 1 cover a pressure range up to 1051 MPa.

Finally, the isobaric ideal-gas heat capacity employed by Span and Wagner⁶ was obtained from Jaeschke and Schley⁵³

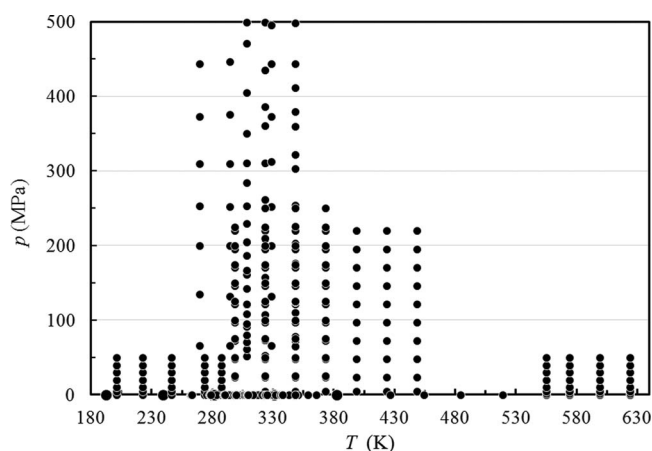


Fig. 1. Temperature and pressure ranges of the primary experimental thermal conductivity data for *n*-heptane.

for the range 200–1000 K. The quoted uncertainty is on the order of 0.05% outside the critical region. We also adopt the values for the critical point and triple point from their EOS; the critical temperature, T_c , and the critical density, ρ_c , were taken to be equal to 540.13 K, and 232.00 kg m^{-3} , respectively.⁶ The triple-point temperature is 182.6 K.⁶

3.1. The dilute-gas limit

From the primary measurements shown in Table 1, only one investigator¹¹ performed measurements near the dilute-gas limit. To increase the temperature range covered, measurements of five more investigators,^{26,31,39,43,47} initially classified as secondary data, were also included. No measurements are ever performed at zero pressure and the values of thermal conductivity are usually obtained by extrapolating the data along an isotherm to a dilute-gas limit. Only Tarzimanov and Masirov³¹ studied the influence of pressure on thermal conductivity of *n*-heptane in the vapor region. Examination of their data indicates that the variation of the thermal conductivity with pressure is slight and decreases with increasing temperature. In this case, we have extrapolated their data to zero density. All other investigators performed the measurements at sufficiently low pressures that the estimated zero-pressure correction would be smaller than 0.5%, and hence we have used their quoted values as the dilute-gas thermal conductivity and marginally increased their claimed uncertainty.

In order to be able to extrapolate the temperature range of the aforementioned measurements (300–650 K), a theoretically based scheme was preferred in order to correlate the dilute-gas limit thermal conductivity, $\lambda_o(T)$, over a wide temperature range. In our previous work,^{1–5} the scheme chosen was based on estimating the thermal conductivity, $\lambda_o(T)$, of the pure dilute gas from its viscosity and ideal-gas heat capacity at constant volume through a modified Eucken correlation.⁵⁴ This scheme worked quite successfully for small spherical-like molecules, but started to produce larger deviations in longer hydrocarbons.⁵ Thus, an alternative approach was adopted in this work.⁵⁵

The traditional kinetic theory approach for thermal conductivity results in an expression involving three generalized cross sections.⁵⁶ However, it is possible to derive an equivalent kinetic theory expression for thermal conductivity by making use of the Thijssse *et al.* approach,^{57,58} where one considers an expansion in terms of total energy, rather than separating translational from internal energy as is done traditionally. In this case, the dilute-gas limit thermal conductivity, $\lambda_o(T)$ ($\text{mW m}^{-1} \text{K}^{-1}$), of a polyatomic gas can be shown to be inversely proportional to a single generalized cross section,^{55–58} $S(10E)$, as

$$\lambda_o(T) = 1000 \frac{5k_B^2(1+r^2)T}{2m\langle v \rangle_o S(10E)} f_\lambda, \quad (2)$$

where k_B is the Boltzmann constant, T (K) is the absolute temperature, $f_\lambda(-)$ is the higher order correction factor, m (kg) is the molecular mass of *n*-heptane ($=0.100202/6.02214 \times 10^{23} \text{ kg}$), and $\langle v \rangle_o = 4\sqrt{k_B T / \pi m}$ (m/s) is the average relative thermal speed. The quantity r^2 is defined by $r^2 = 2C_{\text{int}}^o / 5k_B$, where C_{int}^o is the contribution of both the rotational C_{rot}^o , and the vibrational, C_{vib}^o , degrees of freedom to the isochoric ideal-gas heat capacity C_v^o .

The recent classical trajectory calculations^{59–61} confirm that, for most molecules studied, the higher order thermal conductivity correction factor is near unity. One can take advantage of this finding to define the effective generalized cross section $S_\lambda (=S(10E)/f_\lambda)$ (nm^2), and rewrite Eq. (2) for the dilute-gas limit thermal conductivity of *n*-heptane, $\lambda_o(T)$ ($\text{mW m}^{-1} \text{K}^{-1}$), as

$$\lambda_o(T) = 0.055729 \frac{(C_P^o/k_B) \sqrt{T}}{S_\lambda}. \quad (3)$$

The ideal gas isobaric heat capacity of *n*-heptane, $C_P^o (=C_{\text{int}}^o + 2.5 k_B)$ can be obtained from^{6,53}

$$\begin{aligned} \frac{C_P^o}{k_B} = & 4 + 13.7266 \left(\frac{169.789/T}{\sinh(169.789/T)} \right)^2 \\ & + 30.4707 \left(\frac{836.195/T}{\cosh(836.195/T)} \right)^2 \\ & + 43.5561 \left(\frac{1760.46/T}{\sinh(1760.46/T)} \right)^2. \end{aligned} \quad (4)$$

It has been previously noted,⁵⁷ and recently confirmed⁵⁵ for smaller molecules, that the cross section $S(10E)$ exhibits a nearly linear dependence on the inverse temperature. Preliminary examination of the available experimental data for *n*-heptane indicates that below 440 K there is a general agreement of the $S(10E)$ values, within $\pm 2\%$ – 3% , among different authors. At high temperatures, the three available measurement sets exhibit systematic differences which cannot be reconciled easily. In order to address this, we initially used the data up to 440 K to fit the effective cross section S_λ (nm^2) linearly to inverse temperature. The extrapolation of this expression to higher temperatures indicated that the data of Naziev *et al.*¹¹ have a different temperature dependence than all the other data considered. It is interesting to note that similar behavior was observed⁵ for

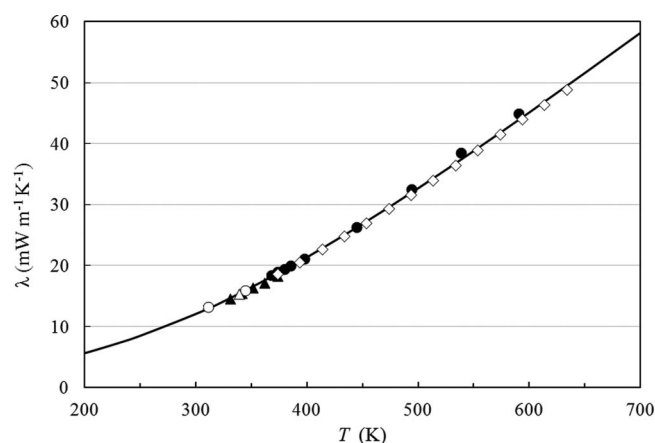


FIG. 2. Dilute-gas limit thermal conductivity as a function of temperature. Shashkov *et al.*²⁶ (▲), Tarzimanov and Mashirov³¹ (●), Carmichael and Sage³⁹ (○), Golubev and Naziev⁴³ (◇), Lambert *et al.*⁴⁷ (Δ), Eq. (6) (—).

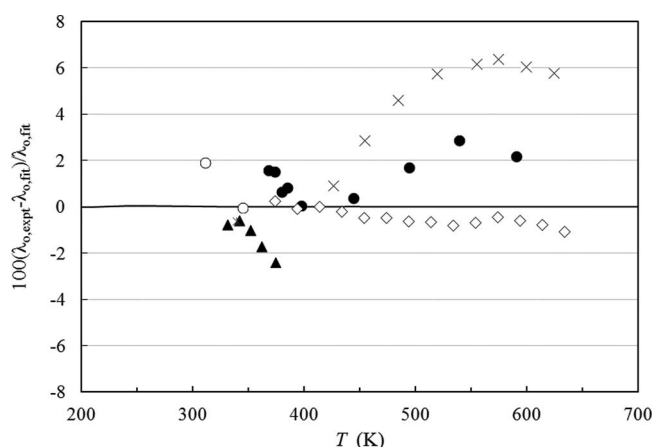


FIG. 3. Percentage deviations of primary experimental data for dilute-gas limit thermal conductivity of *n*-heptane from the values calculated by Eqs. (3)–(5). Naziev *et al.*¹¹ (×), Shashkov *et al.*²⁶ (▲), Tarzimanov and Mashirov³¹ (●), Carmichael and Sage³⁹ (○), Golubev and Naziev⁴³ (◇), Lambert *et al.*⁴⁷ (Δ), Eq. (6) (—).

Naziev *et al.* data for *n*-hexane.¹¹ There are no indications in Ref. 11 why there is a systematic difference between the data of Naziev *et al.*¹¹ and those of Golubev and Naziev.⁴³ We consequently did not further consider the data of Naziev *et al.*¹¹ for the development of the dilute-gas limit thermal conductivity correlation.

In order to develop the correlation, we fitted the effective cross section S_λ (nm²), obtained from experimental data,^{26,31,39,43,47} weighted by uncertainty, by means of Eq. (3) to a polynomial in inverse temperature, resulting in the following expression:

$$S_\lambda = 0.4938 + 330.31/T. \quad (5)$$

Hence, Eqs. (3)–(5) form a consistent set of equations for the calculation of the dilute-gas limit thermal conductivity of *n*-heptane. The experimental dilute-limit thermal-conductivity values as well as the values calculated by Eqs. (3)–(5) are shown in Fig. 2, while Fig. 3 presents the percentage deviations of the dilute-gas experimental data from the values calculated by Eqs. (3)–(5). All the selected data are represented to within $\pm 3\%$, which is commensurate with the uncertainty of the data. No obvious systematic trends are observed. The data of Shashkov *et al.*²⁶ show a similar trend to that already observed for their *n*-hexane data with deviations increasing with increasing temperature. The data of Naziev *et al.*¹¹ are also included only to illustrate the systematic differences observed.

The values of the dilute-gas limit thermal conductivity, $\lambda_o(T)$, in mW m^{−1} K^{−1}, obtained by the scheme of Eqs. (3)–(5), were fitted as a function of the reduced temperature, $T_r = T/T_c$, for ease of use,

Values calculated by Eq. (6) do not deviate from the values calculated by the scheme of Eqs. (3)–(5) by more than 0.1% over the temperature range 182–1000 K. This equation is hence employed in the calculations that will follow.

Based upon the aforementioned discussion, Eqs. (3)–(5), or Eq. (6), represent the dilute-gas limit thermal conductivity to within 2.4% at the 95% confidence level.

3.2. The residual thermal conductivity

The thermal conductivities of pure fluids exhibit an enhancement over a large range of densities and temperatures around the critical point and become infinite at the critical point. This behavior can be described by models that produce a smooth crossover from the singular behavior of the thermal conductivity asymptotically close to the critical point to the residual values far away from the critical point.^{62–64} The density-dependent terms for thermal conductivity can be grouped according to Eq. (1) as $[\Delta\lambda(\rho, T) + \Delta\lambda_c(\rho, T)]$. To assess the critical enhancement either theoretically or empirically, we need to evaluate, in addition to the dilute-gas thermal conductivity, the residual thermal-conductivity contribution. The procedure adopted during this analysis used ODRPACK (Ref. 65) to fit all the primary data simultaneously to the residual thermal conductivity and the critical enhancement, while maintaining the values of the dilute-gas thermal-conductivity data obtained by Eq. (6). The density values employed were obtained by the EOS of Span and Wagner.⁶

The residual thermal conductivity was represented with a polynomial in reduced temperature, T_r , and reduced density,

$$\lambda_o(T) = \frac{-1.83367 + 16.2572 T_r - 39.0996 T_r^2 + 47.8594 T_r^3 + 15.1925 T_r^4 - 3.39115 T_r^5}{0.250611 - 0.320871 T_r + T_r^2}. \quad (6)$$

TABLE 2. Coefficients of Eq. (7) for the residual thermal conductivity of *n*-heptane

<i>i</i>	$B_{1,i}$ (W m ⁻¹ K ⁻¹)	$B_{2,i}$ (W m ⁻¹ K ⁻¹)
1	$5.177\ 85 \times 10^{-2}$	$-7.724\ 33 \times 10^{-3}$
2	$-9.240\ 52 \times 10^{-2}$	$2.188\ 99 \times 10^{-2}$
3	$5.114\ 84 \times 10^{-2}$	$1.717\ 25 \times 10^{-3}$
4	$-7.768\ 96 \times 10^{-3}$	$-7.916\ 42 \times 10^{-3}$
5	$1.216\ 37 \times 10^{-4}$	$1.833\ 79 \times 10^{-3}$

$\rho_r = \rho/\rho_c$, as

$$\Delta\lambda(\rho, T) = \sum_{i=1}^5 (B_{1,i} + B_{2,i}T_r)(\rho_r)^i. \quad (7)$$

The coefficients $B_{1,i}$ and $B_{2,i}$ are shown in Table 2.

3.3. The critical enhancement

3.3.1. Simplified crossover model

The theoretically based crossover model proposed by Olchowy and Sengers^{62–64} is complex and requires solution of a quartic system of equations in terms of complex variables. A simplified crossover model has also been proposed by Olchowy and Sengers.⁶⁶ The critical enhancement of the thermal conductivity from this simplified model is given by

$$\Delta\lambda_c = \frac{\rho C_p R_D k_B T}{6\pi\bar{\eta}\xi} (\bar{\Omega} - \bar{\Omega}_0) \quad (8)$$

with

$$\bar{\Omega} = \frac{2}{\pi} \left[\left(\frac{C_p - C_v}{C_p} \right) \arctan(\bar{q}_D \xi) + \frac{C_v}{C_p} \bar{q}_D \xi \right] \quad (9)$$

and

$$\bar{\Omega}_0 = \frac{2}{\pi} \left[1 - \exp \left(- \frac{1}{(\bar{q}_D \xi)^{-1} + (\bar{q}_D \xi \rho_c / \rho)^2 / 3} \right) \right]. \quad (10)$$

In Eqs. (8)–(10), k_B is Boltzmann's constant, $\bar{\eta}$ is the viscosity, and C_p and C_v are the isobaric and isochoric specific heat obtained from Span and Wagner.⁶ To estimate the viscosity, an unpublished correlation⁶⁷ implemented in the REFPROP (Ref. 68) program was employed. The correlation length ξ is given by

$$\xi = \xi_0 \left(\frac{p_c \rho}{\Gamma \rho_c^2} \right)^{\nu/\gamma} \left[\frac{\partial \rho(T, \rho)}{\partial p} \right]_T - \left(\frac{T_{\text{ref}}}{T} \right) \frac{\partial \rho(T_{\text{ref}}, \rho)}{\partial p} \Big|_T \Big]^{1/\gamma}. \quad (11)$$

As already mentioned, the coefficients $B_{1,i}$ and $B_{2,i}$ in Eq. (7) and $\bar{q}_D$ in Eqs. (8)–(11) were fitted with ODRPACK (Ref. 65) to the primary data for the thermal conductivity of *n*-heptane. This crossover model requires the universal constants⁶⁶ $R_D = 1.02$, $\nu = 0.63$, and $\gamma = 1.239$, and system-dependent amplitudes Γ and ξ_0 . For this work we adopted the values $\Gamma = 0.0586$ and estimated $\xi_0 = 2.45 \times 10^{-10}$ m, using the method

TABLE 3. Evaluation of the *n*-heptane thermal-conductivity correlation for the primary data

First author	Year publ.	AAD (%)	BIAS (%)
Watanabe ⁸	2002	1.25	1.25
Tanaka ⁹	1988	1.33	-1.24
Assael ¹⁰	1987	0.72	-0.72
Naziev ¹¹	1984	2.95	0.09
Forsman ¹²	1982	1.33	0.59
Kashiwagi ¹³	1982	1.42	-1.42
Menashe ¹⁴	1981	0.64	0.34
Nagasaka ¹⁵	1981	0.50	0.50
Naziev ¹⁶	1981	3.29	3.29
Nieto de Castro ¹⁷	1976	1.11	-1.11
Kandiyoti ¹⁸	1972	0.52	-0.32
Mukhamedzyanov ¹⁹	1971	1.71	-0.84
Entire data set		1.49	0.07

presented by Perkins *et al.*⁶⁹ The effective cutoff wavelength $\bar{q}_D^{-1}$ was found equal to 8.0×10^{-10} m. The reference temperature T_{ref} , far above the critical temperature where the critical enhancement is negligible, was calculated by $T_{\text{ref}} = (3/2) T_c$, which for *n*-heptane is 810.2 K.

Table 3 summarizes comparisons of the primary data with the correlation. We have defined the percent deviation as $\text{PCTDEV} = 100 * (\lambda_{\text{exp}} - \lambda_{\text{fit}}) / \lambda_{\text{fit}}$, where λ_{exp} is the experimental value of the thermal conductivity and λ_{fit} is the value calculated from the correlation. Thus, the average absolute percent deviation (AAD) is found with the expression $\text{AAD} = (\sum |\text{PCTDEV}|) / n$, where the summation is over all n points. The bias percent is found with the expression $\text{BIAS} = (\sum \text{PCTDEV}) / n$. The standard deviation of the overall fit is 2%.

Figure 4 shows the percentage deviations of all primary thermal-conductivity data from the values calculated by Eqs. (1) and (6)–(11), as a function of the density, while Figs. 5

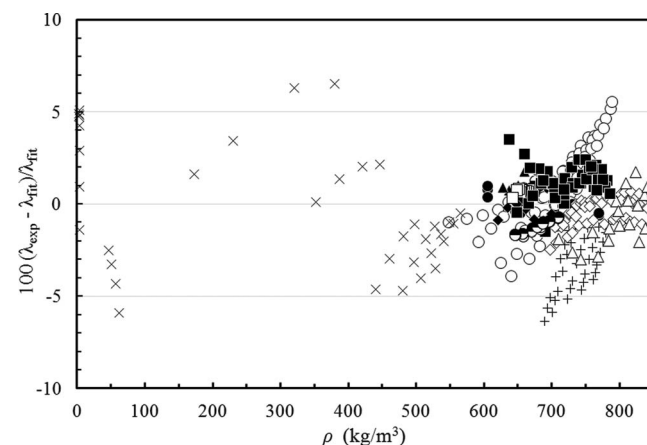


FIG. 4. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of density. Watanabe and Seong⁸ (●), Tanaka *et al.*⁹ (■), Assael *et al.*¹⁰ (□), Naziev *et al.*¹¹ (×), Forsman *et al.*¹² (Δ), Kashiwagi *et al.*¹³ (▲), Menashe and Wakeham¹⁴ (◇), Nagasaka and Nagashima¹⁵ (◆), Naziev *et al.*¹⁶ (+), Nieto de Castro *et al.*¹⁷ (●), Kandiyoti *et al.*¹⁸ (●), Mukhamedzyanov *et al.*¹⁹ (○).

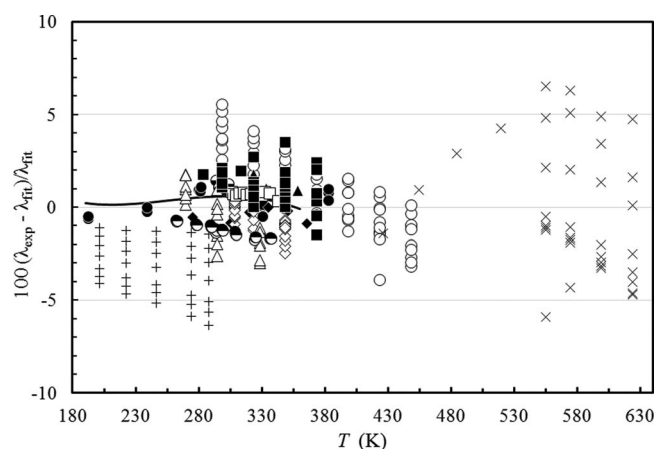


Fig. 5. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature. Watanabe and Seong⁸ (●), Tanaka *et al.*⁹ (■), Assael *et al.*¹⁰ (□), Naziev *et al.*¹¹ (×), Forsman *et al.*¹² (Δ), Kashiwagi *et al.*¹³ (▲), Menashe and Wakeham¹⁴ (◇), Nagasaka and Nagashima¹⁵ (◆), Naziev *et al.*¹⁶ (+), Nieto de Castro *et al.*¹⁷ (●), Kandiyoti *et al.*¹⁸ (●), Mukhamedzyanov *et al.*¹⁹ (○), Eq. (12) (—).

and 6 show the same deviations but as a function of the temperature and pressure. The primary data for *n*-heptane listed in Table 1 cover a wide range of conditions and extend to 1051 MPa. As mentioned earlier, the EOS of Span and Wagner⁶ used here for density can be applied up to 250 MPa pressure. Based on comparisons with the primary data, we calculate the uncertainty (at the 95% confidence level) for pressures less than 250 MPa and temperatures less than 600 K to be less than 4%. Uncertainties in the critical region are much larger, since the thermal conductivity approaches infinity at the critical point and is very sensitive to small changes in density. Examining Fig. 6, it seems that the correlation can be extended to 500 MPa without any increase of uncertainty—densities were estimated by extrapolating the values of Span and Wagner.⁶

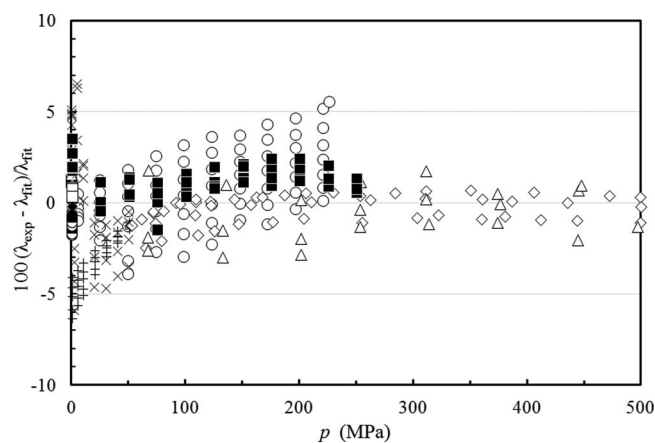


Fig. 6. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of pressure. Watanabe and Seong⁸ (●), Tanaka *et al.*⁹ (■), Assael *et al.*¹⁰ (□), Naziev *et al.*¹¹ (×), Forsman *et al.*¹² (Δ), Kashiwagi *et al.*¹³ (▲), Menashe and Wakeham¹⁴ (◇), Nagasaka and Nagashima¹⁵ (◆), Naziev *et al.*¹⁶ (+), Nieto de Castro *et al.*¹⁷ (●), Kandiyoti *et al.*¹⁸ (●), Mukhamedzyanov *et al.*¹⁹ (○).

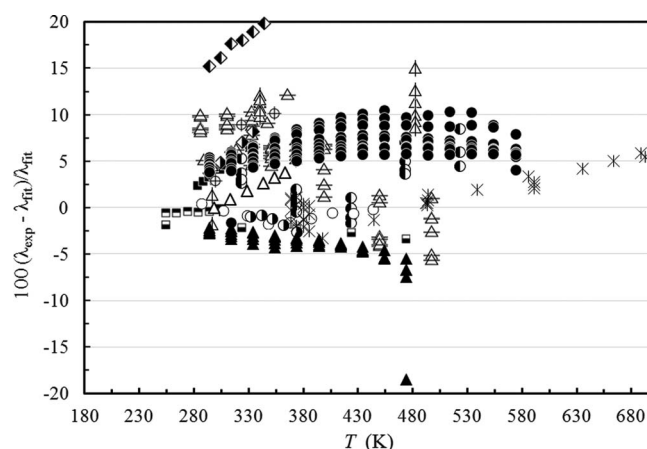


Fig. 7. Percentage deviations of secondary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature (period 1969–1997). Lei *et al.*²⁰ (♦), Tong and Li²¹ (●), Rowley *et al.*²² (+), Knibbe and Raal²³ (○), Fischer and Ernfried²⁴ (■), El-Sharkawy *et al.*²⁵ (◆), Shashkov *et al.*²⁶ (●), Magerramov²⁷ (▲), Atalla *et al.*²⁸ (■), Guseinov and Klimova²⁹ (Δ), Ogiwara *et al.*³⁰ (◆), Tarzimanov and Mashirov³¹ (×), Mallan *et al.*³² (Δ), Naziev *et al.*³³ (●), Kerimov *et al.*³⁴ (◆), Bogatov³⁵ (▲).

In 1986, Nieto de Castro *et al.*⁷⁰ proposed the following correlation for the thermal conductivity, λ_{sat} (mW m⁻¹ K⁻¹), of *n*-heptane along the liquid saturation line

$$\lambda_{\text{sat}}(T) = 212.475 - 89.635(T/298.15), \quad (12)$$

valid from 191 to 365 K with an uncertainty of 1.5%, as a secondary reference correlation. The values calculated by the present scheme, which is based upon a larger number of investigators, agree with the correlation of Eq. (12) within 0.3% as can be seen in Fig. 5.

Figures 7 and 8 show the percentage deviations of the secondary data from the values calculated by the proposed scheme. The majority of the deviations, except some very old sets, are within 5% of the present correlation.

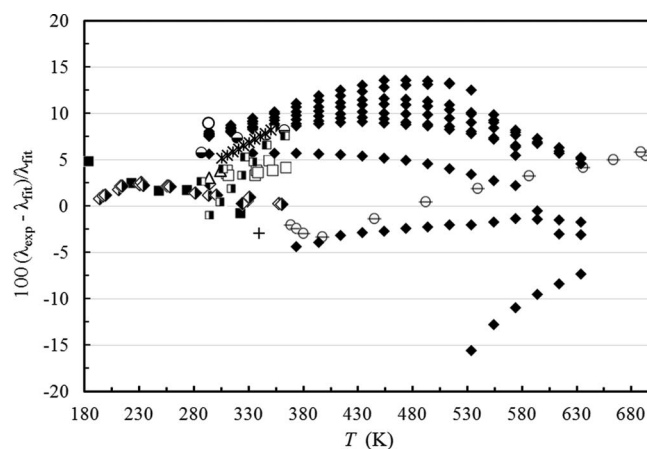


Fig. 8. Percentage deviations of secondary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature (period 1913–1968). Brykov *et al.*³⁶ (◆), Abas-Zade and Guseinov³⁷ (●), Tarzimanov and Mashirov³⁸ (○), Carmichael and Sage³⁹ (●), Jobst⁴⁰ (■), Akhmedov⁴¹ (◆), Mukhamedzyanov *et al.*⁴² (□), Golubev and Naziev⁴³ (◆), Vilim⁴⁴ (○), Frontasev and Gusakov⁴⁵ (Δ), Briggs⁴⁶ (■), Lambert *et al.*⁴⁷ (+), Sakiadis and Coates⁴⁸ (×), Filippov⁴⁹ (■), Hernqvist⁵⁰ (▲).

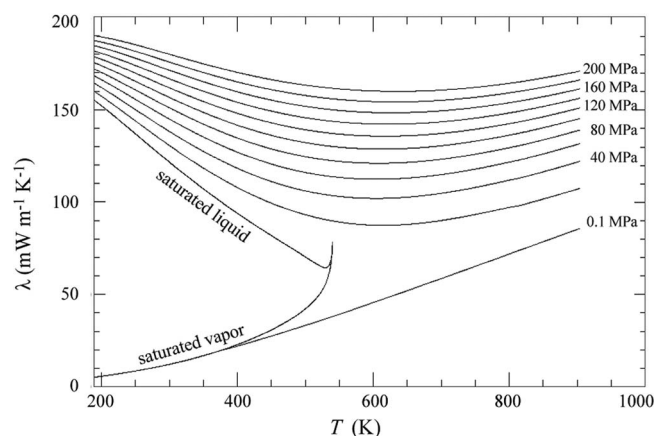


FIG. 9. Thermal conductivity of *n*-heptane as a function of temperature for different pressures.

Finally, Fig. 9 shows a plot of the thermal conductivity of *n*-heptane as a function of the temperature for different pressures.

3.3.2. Empirical critical enhancement

For applications at state points that are relatively distant from the critical point (at least 10–15 K from the critical temperature), the critical enhancement is adequately represented by the following empirical expression:

$$\Delta\lambda_c(\rho, T) = \frac{C_1}{C_2 + |\Delta T_c|} \exp[-(C_3 \Delta\rho_c)^2], \quad (13)$$

where $\Delta T_c = (T/T_c) - 1$ and $\Delta\rho_c = (\rho/\rho_c) - 1$. This equation does not require accurate information on the compressibility, specific heat, and viscosity of *n*-heptane in the critical region, as does the theory of Olchowy and Sengers.^{63,64,66} The coefficients of Eqs. (6) and (7) were fixed, while the coefficients of

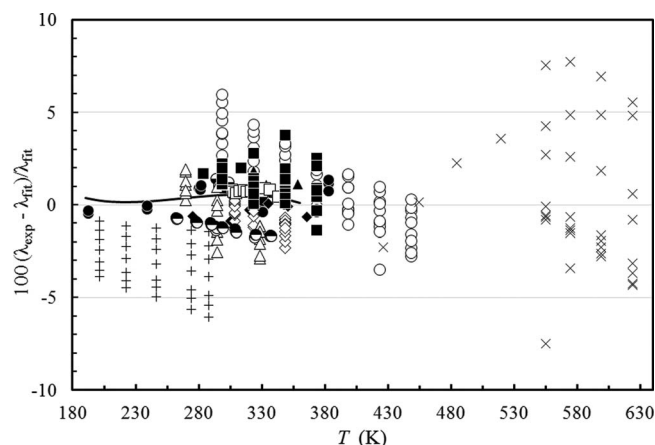


FIG. 10. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by Eqs. (1), (6), and (7) and the empirical enhancement of Eq. (13) as a function of temperature. Watanabe and Seong⁸ (●), Tanaka *et al.*⁹ (■), Assael *et al.*¹⁰ (□), Naziev *et al.*¹¹ (×), Forsman *et al.*¹² (Δ), Kashiwagi *et al.*¹³ (▲), Menashe and Wakeham¹⁴ (◇), Nagasaka and Nagashima¹⁵ (◆), Naziev *et al.*¹⁶ (+), Nieto de Castro *et al.*¹⁷ (●), Kandiyoti *et al.*¹⁸ (●), Mukhamedzyanov *et al.*¹⁹ (○), Eq. (12) (—).

TABLE 4. Sample points for computer verification of the correlating equations

T (K)	ρ (kg m ⁻³)	λ (mW m ⁻¹ K ⁻¹)
250.00	720.00	137.09
400.00	2.00	21.794
400.00	650.00	120.75
535.00	100.00	51.655 ^a
535.00	100.00	49.681 ^b

^aComputed with modified Olchowy–Sengers critical enhancement; the viscosity at this point for use in Eq. (8) was taken as $\eta = 15.253 \mu\text{Pa s}$ (see Sec. 3.3.1); all other properties required for the enhancement term are from Span and Wagner⁶.

^bComputed with empirical critical enhancement Eq. (13).

Eq. (13) were fitted to the primary data. The values obtained were $C_1 = 0.7 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$, $C_2 = 7.0 \times 10^{-2}$, and $C_3 = 1.8$. Figure 10 shows the percentage deviations between the primary data and the values calculated by Eqs. (1), (6), (7), and (13), as a function of the temperature. By comparing Figs. 5 and 10, it can be seen that employing Eq. (13) results in very little deterioration in the representation of the data.

4. Computer-Program Verification

Table 4 is provided to assist the user in computer-program verification. The thermal-conductivity calculations are based on the tabulated temperatures and densities. Note that Eq. (6) was employed for the dilute-gas correlation.

5. Conclusion

New wide-ranging correlations for the thermal conductivity of *n*-heptane were developed based on critically evaluated experimental data. The correlations are valid from the triple point to 600 K, and at pressures up to 250 MPa. The correlations are expressed in terms of temperature and density, and the densities were obtained from the EOS of Span and Wagner.⁶ The overall uncertainty (at the 95% confidence level) of the proposed correlation is estimated, for pressures up to 250 MPa and temperatures less than 600 K, to be less than 4%. Moreover, it seems that the correlation can be extended to 500 MPa without a significant increase in uncertainty.

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