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PREDICTION OF THE THERMAL CONDUCTIVITY OF SATURATED LIQUIDS

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ABSTRACT

Using the dimensionless coordinates

$$\lambda^* = \left(\frac{\lambda_c}{\lambda} - 1\right) / \left(\frac{\lambda_c}{\lambda_{NB}} - 1\right),$$

where λ_c = thermal conductivity at the critical point, λ_{NB} = thermal conductivity at the normal boiling temperature, T_{NB} (K), and λ = thermal conductivity at any given temperature, T(K) and

$$T^* = \left(\frac{T_c}{T} - 1\right) / \left(\frac{T_c}{T_{NB}} - 1\right),$$

where T_c = the critical point temperature (K); an empirical equation,

$$\lambda^* = \left[\frac{mT^*}{k + T^*}\right]^b,$$

has been established to calculate the thermal conductivity of saturated liquids over the entire saturated liquid–vapour equilibrium range from the triple point to the critical point for separate compounds as well as the C_1–C_{10} alkanes, inert and halogen compound families. The relation proposed in this work compares excellently in overall accuracy with other known selected prediction correlations over the entire range of investigation (25 compounds with 548 saturated liquid-state data points).

INTRODUCTION AND PROPOSED CORRELATION

The need for predicting the thermal conductivity of saturated liquids as a function of saturation temperature stems first, from the overall paucity of the available experimental data [1], and second, from the current advances in chemical engineering design, especially heat-transfer operations. To remedy this need, a flurry of theoretical and semi-empirical thermal conductivity prediction methods for liquids has been proposed [2].

However, these proposals have met with indifferent success. The theoretical ones have not checked out very well, while the semi-empirical predictive

TABLE 1

Coefficients for use in eqn. (4)

Compound	Original coefficients			Truncated coefficients		
	m	*k*	*b*	*m*	*k*	*b*
Ammonia	1.311886559184984	0.3093307000967518	1.019198160032136	1.3118	0.3093	1.0191
Argon	2.117636829650496	1.105929540812912	0.1919117514815389			
Bromine	1.203719961044353	0.2117571926252732	0.4168803501749347			
n-Butane	4.533661487574754	3.515620537724174	0.4583262104259282			
Chlorine	1.483561590011864	0.4877431511875799	0.2818616184984869			
n-Decane	0.4489513741093500	−0.5702676434818911	−0.33080915656432679			
Ethane	2.206579022481452	1.237164310042794	0.4457849293668946			
Ethylene	1.146647660415077	0.1393321311046534	1.607944516858232	1.1466	0.1393	1.6079
Fluorine	1.640513501757393	0.6297100018214363	0.2743923961466103			
n-Heptane	1.997924145463241	0.9950788160501393	0.8710986893865686			
n-Hexane	1.801493598024241	0.8040697207870712	1.198115338275912			
Iodine	2.620093282372702	1.556222778147153	0.1989802932396336			
Krypton	1.413676368082480	0.4191643272078135	0.3681386240455116			
Methane	1.844655825917703	0.8740434110085401	0.4865832476024233			
Neon	1.272068578900701	0.2452914648895645	0.3895389875399014			
Nitric oxide	1.619786783298031	0.6203536697827889	0.2717658515655353	1.6197	0.6203	0.2717
Nitrogen	1.407252593535153	0.4069618816256084	0.2806466925007127	1.4072	0.4069	0.2806

n-Nonane	1.759556700743059	0.7609251378559813	0.7443024534297075			
n-Octane	1.311549544048340	0.3140015693802816	1.631432762499684			
Oxygen	1.441284534592610	0.4487318064065133	0.3879387949128753	1.4412	0.4487	0.3879
n-Pentane	1.731740736839991	0.7499948951897793	1.171024035514059			
Propane	1.251117637459089	0.2742182904484753	0.9977451712306740			
Radon	1.555545369659313	0.5662807061607264	0.2833095579765076			
Water[a]						
Xenon	1.505967428982191	0.5191892127825420	0.3107704086624491			

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[a] The curve for water has been broken up into three temperature ranges. The range 403.15 $< T < 633.15$ K is represented by the hyperbolic equation

$$\lambda^* = m + \frac{k}{T^* + b}$$

The two remaining regions are represented by eqn. (4). The coefficients are as follows

Temperature range (K)	Original coefficients			Truncated coefficients		
	m	k	b	m	k	b
$T < 403.15$	6.810171411730729	5.99999999905579	0.4156490503755682	6.8102	0.4156	6.0000
$403.15 < T < 633.15$	0.8164286414636723	0.1367055701032584	0.1136019627899926	0.8164	0.1367	0.1136
$633.15 < T$	7.267108182181715	0.00062442064306273875	0.3559901138517853	7.2671	0.0006244	0.3560

methods at times give very good results, but then, again, yield estimates subject to an excessive error [2].

Despite the relatively early studies of the changes in thermal conductivity with temperature along the saturated liquid–vapour equilibrium curve [3,4], to date there is no satisfactory method for predicting the thermal conductivity of saturated liquids.

Since these saturated equilibrium curves represent states of coexistence of pairs of phases (i.e., liquid–vapour) and the system is thermodynamically univariant, we can say that for a pure substance at a given temperature T and for a given value of $\lambda(T)$, a single value of P exists, satisfying the following thermodynamic restriction

$$\lambda_s = \mathrm{g}(T_s) = \mathrm{h}(P_s) \tag{1}$$

where subscript s indicates the saturated state with g and h representing functions. Equation (1) specifies that only either the saturation temperature or pressure is necessary to describe the thermal conductivity of the saturated liquid uniquely, since choosing T_s will at the same time determine the value of P_s for the given substance. In view of eqn. (1) it is feasible to develop a direct empirical relation between the saturated liquid thermal conductivity and the temperature.

To establish such a relation, we introduce [5–8] new reduced coordinates associated with the phenomenological scaling renormalization group theory [9–12] as follows

$$\lambda^* = \left(\frac{\lambda_c}{\lambda} - 1\right) \Big/ \left(\frac{\lambda_c}{\lambda_{NB}} - 1\right) \tag{2}$$

and

$$T^* = \left(\frac{T_c}{T} - 1\right) \Big/ \left(\frac{T_c}{T_{NB}} - 1\right) \tag{3}$$

where λ_c = thermal conductivity at the critical point; λ_{NB} = thermal conductivity at the normal boiling point temperature, T_{NB}(K); λ = thermal conductivity at any given temperature, T(K); and T_c = critical point temperature (K).

For this coordinate system (eqns. 2 and 3) the thermal conductivity of the saturated liquid of a pure substance can then be expressed as follows (Tables 1 and 2, Fig. 1)

$$\lambda^* = \left[\frac{mT^*}{k + T^*}\right]^b \tag{4}$$

where m, b and k are constants characteristic of the pure substance in its saturated liquid state, and have been obtained from experimental data sets (Table 3) by means of non-linear least-square regression methods.

TABLE 2

Coefficient-MW expansion (eqn. 5) of C_1–C_{10} alkane, halogen and inert liquid families for use in eqn. (4)

Compound family	Original coefficient expansion of eqn. (5)	Truncated coefficient expansion of eqn. (5)
C_1–C_{10} alkanes	$m = -1.827067528701\text{E-}04\ (\mathrm{MW} - 89.8332429388)^2 + 1.93649315343$	$m = -1.8270\text{E-}04\ (\mathrm{MW} - 89.83)^2 + 1.936$
	$k = -1.552282535611\text{E-}04\ (\mathrm{MW} - 91.6153377467)^2 + 0.901276329364$	$k = -1.5522\text{E-}04\ (\mathrm{MW} - 91.61)^2 + 0.9012$
	$b = 0.89135829189322$	$b = 0.8913$
Halogens	$m = -1.3674980430361582\text{E-}06\ (\mathrm{MW} + 106.9742791349550)^2 + 1.541884606235886$	$m = -1.3674\text{E-}06\ (\mathrm{MW} + 106.9)^2 + 1.541$
	$k = -5.4357643124079702\text{E-}04\ (\mathrm{MW} + 0.5275404439183262)$	$k = -5.4357\text{E-}04\ (\mathrm{MW} + 0.5275)$
	$b = 0.2930286645149163\,26$	$b = 0.2930$
Inerts	$m = -1.8986040534591662\text{E-}05\ (\mathrm{MW} - 130.2408309945388)^2 + 1.578245861226605$	$m = -1.8986\text{E-}05\ (\mathrm{MW} - 130.2)^2 + 1.578$
	$k = -1.6387300924175560\text{E-}05\ (\mathrm{MW} - 139.6353743273155)^2 + 0.5555561193558613$	$k = -1.6387\text{E-}05\ (\mathrm{MW} - 139.6)^2 + 0.5556$
	$b = 0.3087338659411816\,57$	$b = 0.3087$

The proposed method (eqn. 4) was generalized in terms of the given compound families. In this work we could propose generalized correlations for the C_1–C_{10} liquid alkane, liquid inert and liquid halogen families (Table 2, Fig. 1) by noting that the m and k coefficients of eqn. (4) can be expressed as non-linear functions of molecular weight

$$X = P(1)[\mathrm{MW} - P(2)]^2 + P(3) \qquad (5)$$

($k = P(1)\mathrm{MW} - P(2)$ for liquid halogens) where X is the running variable for the m and k parameters, while the coefficients $P(1)$, $P(2)$ and $P(3)$, determined by means of non-linear regression techniques, are shown both in their long and truncated forms in Table 2. The m and k functions (eqn. 5) are both family as well as state dependent, while b remains a constant for the particular compound family and the liquid state.

The validity of the correlation presented was tested with excellent success by comparing the RMS% error values of this work against the RMS% error values as obtained in combination of the Boiling Point–Robbins–Kingrea equation [2,13] and the Missenard equation [14] for the low-pressure range, and then extending these results to high pressures by the use of the Lenoir correlation [15].

EXPERIMENTAL DATA USED

A summary of experimental data used, along with their temperature ranges, is given in Table 3. These saturated liquid thermal conductivity data

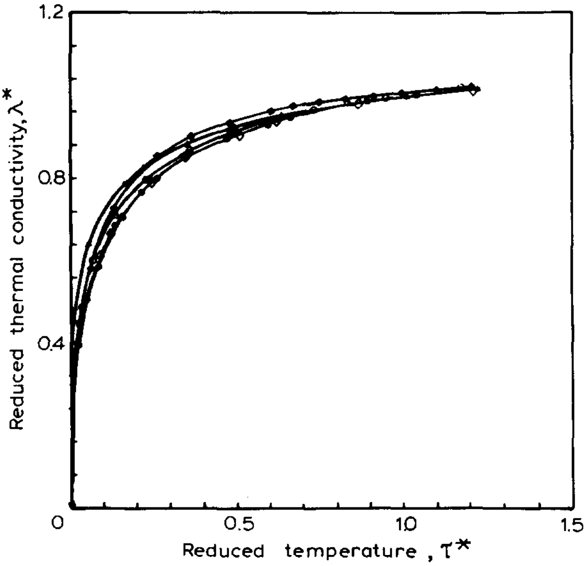

Fig. 1. Reduced thermal conductivity (λ*) vs. reduced temperature (T^*) plot: (◆) neon; (▲) argon; (●) krypton; (◇) xenon; (○) radon.

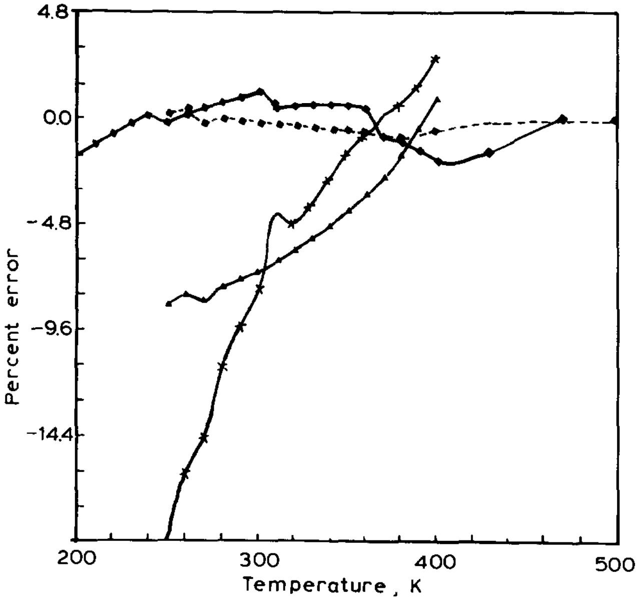

Fig. 2. Percent error in thermal conductivity vs. temperature plot for *n*-heptane: (◆———◆) calculated using eqn. (5), original coefficients; (◆------◆) calculated using eqn. (4), original coefficients; (▲———▲) calculated by means of Boiling Point–Robbins–Kingrea equation; (×———×) calculated by means of Missenard equation.

were taken from several sources. Table 4 presents the experimental data needed for use in both the Boiling Point–Robbins–Kingrea equation [2,13] and the Missenard equation [14]. One is directed to the paper by Lenoir [15] for the correlation diagram used in adjusting thermal conductivity for pressure effects. The data presented (Tables 3 and 4) were considered to be sufficiently accurate and no further studies were made to re-evaluate their accuracy. Hence, the presently used data sets (Tables 3 and 4) are only representative ones. The RMS% error is used as a basis for comparison thus facilitating a measure of accuracy if fit for this work (eqns. 4 and 5) and other methods (Table 5, Fig. 2).

RESULTS AND DISCUSSION

The introduction of the proposed λ^*-T^* relation (eqn. 4) enables one to calculate the saturated-liquid thermal conductivities for pure compounds along the entire range of the saturated liquid–vapour equilibrium curve from the triple point up to the critical point.

The dimensionless parameter λ^*-T^* relation (eqns. 4 and 5) associated with the phenomenological scaling and group renormalization theory [10,12,16] introduced in this work, eliminates the need for any direct

TABLE 3

Summary of experimental data used

Compound (literature cited)	Normal boiling point temp. (K)	Critical point temp. (K)	Liquid thermal conductivity at normal boiling point (W m^{-1} K^{-1})	Thermal conductivity at critical point (W m^{-1} K^{-1})	No. of data points for liquid	Molecular weight	Reduced temp. range (liquid)
Ammonia [2,17,18]	240.0	405.5	0.614	0.2425	20	–	0.543–1.000
Argon [18–20]	87.29	151.2	0.1232	0.025	16	39.948	0.529–1.000
Bromine [2,18,19]	331.9	584.0	0.11575	0.0280	35	159.808	0.445–1.000
n-Butane [2,21,22]	272.7	425.31	0.11126	0.05192	46	58.12	0.641–1.000
Chlorine [2,18,19]	238.7	417.0	0.164	0.040	27	70.906	0.408–1.000
n-Decane [18,20,22]	446.0	619.0	0.09037	0.03380	25	142.29	0.371–1.000
Ethane [2,22,23]	184.5	305.4	0.15125	0.05625	26	30.07	0.581–1.000
Ethylene [2,18,24]	170.0	282.4	0.1919	0.07274	20	–	0.352–1.000
Fluorine [2,18,19]	85.0	144.0	0.15758	0.040	12	37.997	0.347–1.000
n-Heptane [18,20,22]	371.0	540.01	0.11020	0.07137	18	100.21	0.463–1.000
n-Hexane	342.0	507.7	0.11278	0.07823	18	86.18	0.492–1.000

Iodine [2,19,25]	457.5	785.0	0.10588	0.02650	24	253.809	0.583–1.000
Krypton [2,18,19]	119.8	210.0	0.09054	0.021	21	83.30	0.548–1.000
Methane [2,18,22]	112.0	190.6	0.19206	0.06615	13	16.06	0.472–1.000
Neon [18–20]	27.09	44.5	0.11234	0.033	22	20.179	0.562–1.000
Nitric oxide [2,25]	121.4	180.0	0.175	0.04949	25	–	0.568–1.000
Nitrogen [2,25]	77.4	126.2	0.13376	0.03579	25	–	0.568–1.000
n-Nonane [18,20,22]	424.0	595.0	0.09796	0.04584	22	128.26	0.420–1.000
n-Octane [18,20,22]	399.0	569.2	0.10325	0.05441	18	114.23	0.439–1.000
Oxygen [18]	90.0	154.7	0.1501	0.041	22	–	0.582–1.000
n-Pentane [18,20,22]	309.0	469.62	0.11456	0.07200	23	72.15	0.426–1.000
Propane [2,22,23]	231.1	369.8	0.09753	0.05538	28	44.11	0.615–1.000
Radon [18,19,22]	211.2	377.2	0.0584	0.0138	20	222.0	0.530–1.000
Xenon [2,19,24]	165.0	289.8	0.07069	0.01683	22	131.30	0.500–1.000
Water [2,20,22,26–28]	373.15	647.30	0.67881	3.65178	44	18.016	0.285–1.000

TABLE 4

Summary of input data for the Missenard and Boiling Point–Robbins–Kingrea equations[a]

Compound (literature cited)	No. of data points	Normal boiling point density (g^{-1} mol^{-1} cm^{-3})	Density at 273 K (g^{-1} mol^{-1} cm^{-3})	Specific heat at normal boiling point (cal g^{-1} mol^{-1} K^{-1})	Specific heat at 273 K (cal g^{-1} mol^{-1} K^{-1})	Critical pressure (ATM)	Reduced temp. range
n-Butane[b] [18,20]	19	0.01034624	0.01033612	31.7682649	31.7867351	34.4636073	0.641–0.754
n-Decane[b] [18,20]	6	0.00425731	0.00523649	77.6470588	72.7287188	20.8931656	0.420–0.501
n-Heptane[b] [18,20]	17	0.00606300	0.0069212	62.0715490	52.2149613	27.0022206	0.463–0.741
n-Hexane[b] [18,20]	6	0.00711714	0.0078579	51.1308980	44.1815167	29.9136442	0.492–0.591
n-Nonane[b] [18,20]	17	0.00473599	0.00571651	67.4545455	65.2512181	22.5808043	0.420–0.521
n-Octane[b] [18,20]	3	0.00535269	0.00628994	70.5779661	58.3864049	24.6336047	0.492–0.527
n-Pentane[b] [18,20]	9	0.00846623	0.0089466	41.6228245	38.2442158	33.2987910	0.426–0.596
Water [2,18,22]	12	0.05319800	0.05550103	8.1312441	7.9925013	217.6	0.422–0.576

[a] T_c, T_{NB} and MW are the same as those in Table 3. Units are such that numbers can be directly used with the equations presented in Reid, Prausnitz and Sherwood [2].

[b] Corrections in high-pressure thermal conductivity were made using the Lenoir correlation, (Reid, Prausnitz and Sherwood [2]). The saturation pressures of ethane (Vargaftik [20]) were placed in reduced coordinates (P_s/P_c) and used to calculate the necessary saturation pressures via the Principle of Corresponding States.

reference to the nature of the saturated liquid–vapour equilibrium, the magnitude and type of the intermolecular forces involved, and the description of the molecular structure of the substance. Instead, the proposed method (eqns. 4 and 5) requires the knowledge of the liquid–vapour critical point and normal boiling point temperatures, along with their corresponding thermal conductivities, and the molecular weight of the compound.

Table 5 compares the results obtained in terms of RMS% error values between the proposed method, original coefficients (eqn. 4, Table 1) and coefficient-molecular weight (MW) expansion (eqn. 5, Table 2), and the Boiling Point–Robbins–Kingrea [2,13] and Missenard [14] equations for the C_1–C_{10} liquid alkanes, liquid inerts and liquid halogens. Since the methods listed are low-pressure correlations, these two test equations were corrected for pressure by means of the Lenoir correlation [15]. Table 5 shows that the method proposed in this work (eqns. 4 and 5), has the smallest RMS% error values among all the test equations selected. Figure 2 illustrates this graphically by giving the plot of RMS% error vs. temperature for liquid *n*-heptane. The proposed method (Table 5) equally well applies to the rare gases, halogens and hydrocarbons (including highly dipolar compounds, such as ammonia). The largest error is found for both the proposed method (eqn. 4) and the other test methods used, for water (Table 5). To increase the ease of using the proposed method (eqns. 4 and 5), the long original coefficients (Tables 1 and 2) for both eqns. (4) and (5), were truncated to coefficients of four decimal places (Tables 1 and 2). Table 5 shows that very little predictive accuracy in the proposed method has been lost as a result of this coefficient truncation.

Methods that work on non-polar organic compounds like the Boiling Point–Robbins–Kingrea equation and the Missenard equation are simply inapplicable to polar compounds like water (Table 5). Although eqn. (5) did not accurately fit the saturated liquid curve for water, reasonable success was obtained in the temperature regions $T < 403.15$ and $T > 633.15$ K (Table 5) with the intermediate temperature range described by a hyperbolic function (Table 5). In general, one can state that the reduced coordinate system of $\lambda^* = f(T^*)$ proved remarkably successful in predicting the liquid thermal conductivities for water.

CONCLUSIONS

A semi-empirical correlation method (eqns. 4 and 5) has been developed which permits one to predict the saturated liquid state thermal conductivity along the entire saturated liquid–vapour equilibrium curve, from the triple point to the critical point with a great deal of accuracy. The characteristic constants of this correlation (m and k for eqns. 4 and 5) have been not only correlated for the individual compounds but, by means of an expansion

TABLE 5

Comparison of results, in RMS% error [a]

Compound	Method proposed in this work				Other methods	
	Coefficient-MW expansion (eqn. 5)		eqn. (4)		Lenoir adjusted Boiling Point–Robbins–Kingrea equation	Lenoir adjusted Missenard equation
	Original *m, k, b,* relations	Truncated *m, k, b* relations	Original *m, k, b* coefficients	Truncated *m, k, b* coefficients		
Ammonia	–	–	0.30	0.29	–	–
Argon	4.39	4.39	0.68	–	–	–
Bromine	1.82	1.81	1.80	–	–	–
Chlorine	1.08	1.06	0.27	–	–	–
Ethane	4.27	4.29	1.35	–	–	–
Ethylene	–	–	2.31	2.31	–	–
Fluorine	1.74	1.76	0.84	–	–	–
Iodine	2.82	2.83	1.54	–	–	–
Krypton	4.01	4.00	0.63	–	–	–
Methane	28.25	28.17	1.88	–	–	–
n-Butane	5.16	5.16	0.73	–	33.75	25.65
n-Decane	4.97	5.03	2.45	–	26.23	1.38
Neon	2.56	2.55	1.41	–	–	–

n-Heptane	0.47	0.46	0.11	–	8.37	5.64
n-Hexane	1.82	1.82	0.25	–	15.44	7.09
Nitric oxide	–	–	0.42	0.42	–	–
Nitrogen	–	–	0.06	0.06	–	–
n-Nonane	2.53	2.49	0.16	–	30.70	1.91
n-Octane	3.83	3.80	0.30	–	9.04	4.32
n-Pentane	2.72	2.72	0.89	–	24.74	10.35
Oxygen	–	–	0.45	0.45	–	–
Propane	2.66	2.67	1.07	–	–	–
Radon	0.94	0.94	0.44	–	–	–
Water	–	–	[b]	[b]	49.69	63.43
Xenon	1.77	1.76	1.13	–	–	–

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[a] Root-mean-square (RMS) percent error

$$\text{RMS\% error} = \left[\frac{\sum_{i=1}^{N} \left(\frac{\text{exp} - \text{calc}}{\text{exp}} \right)_i^2}{\text{No. of points, } N} \right]^{1/2}$$

[b] The curve for water has been broken up into three temperature ranges. The errors are as follows

Temperature range (K)	Original m, k, b coefficients	Truncated m, k, b coefficients
$T < 403.15$	2.61	2.61
$403.15 < T < 633.15$	1.06	1.06
$633.15 < T$	10.37	10.37

through molecular weight (eqn. 5), also as members of the homologous compound series.

The range of application of the proposed method needs to be extended to other compounds, compound families and compound mixtures in saturated and unsaturated states.

The results obtained and comparisons made (Figs. 1 and 2, Table 5) strongly support the proposed method.

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NOMENCLATURE

b, m, k	characteristic coefficients of pure compound, eqn. (4)
$g(T_s)$	arbitrary function of saturation temperature
$h(P_s)$	arbitrary function of saturation pressure
MW	molecular weight
N	number of data points
P	pressure
$P(1)$, $P(2)$, $P(3)$	coefficients in eqn. (5)
T, T^*, T_{NB}, T_c	temperature at any state of the saturated liquid–vapour equilibrium curve; reduced temperature, eqn. (3); normal boiling point temperature; critical point temperature (K), respectively
X	arbitrary (running) variable in eqn. (5)

Subscripts

c	critical point
i	component i, ith data point
NB	normal boiling point
s	saturated state

Superscripts

*	reduced coordinate, eqns. (2) and (3)

Greek letters

λ, λ^*, λ_{NB}, λ_c, λ_s	thermal conductivity at any state; reduced thermal conductivity, eqn. (2); thermal conductivity at the normal boiling point; thermal conductivity at the critical point; thermal conductivity of saturated liquid (W m^{-1} K^{-1}), respectively
$\lambda(T)$	thermal conductivity as a function of temperature

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