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A new model of thermal conductivity for liquids

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Abstract

Using our previous estimation equation of the heat of vaporization for liquids with the residual function method, and with the liquid free volume model as well as the theory of molecular dynamics, a new two-parameter model of thermal conductivity for liquids has been derived. With this new model, the thermal conductivity data for 68 kinds of liquids at 1217 temperature sets were tested. The results are compared with the recent models proposed by Lei et al. [Q. Lie, Y.C. Hou, R. Lin, Chem. Eng. Sci. 52 (1997) 1234; Q. Lie, R. Lin, D. Ni, J. Chem. Eng. Data 42 (1997) 971], Klass et al. [D.M. Klaas, D.S. Viswanath, Ind. Eng. Chem. Res. 37 (1998) 2064], and with the Jamieson equation and the modified Riedel equation. The comparison shows that this model is applicable to many kinds of liquids for a relatively wide range of temperature up to the critical point.

Keywords: Thermodynamics; Modeling; Basic data

1. Introduction

Thermal conductivity data of liquids are important in the design of chemical engineering, but it is difficult to accurately measure them. Even for the same liquid at the same temperature, the experimental thermal conductivity measured by different investigators shows considerable disagreement. Since theoretical methods have not yielded sufficiently accurate and simple expressions for calculating the thermal conductivity of liquids, correlation based on empirical or semi-theoretical method is widely employed over limited ranges of temperature [1–4]. Recent correlations are due to Jamieson [5,6], Teja and co-workers [7–9], Klaas and Viswanath [10], and Lei and co-workers [11,12]. However, many of these methods have some limitations, suggesting that a new model of thermal conductivity for liquids is wanted, which has some theoretical basis and is applicable to more kinds of liquids for a wider range of temperature.

In this paper, a new model of thermal conductivity for liquids, which may be used over wide ranges of temperature, even up to the critical temperature, is proposed on the basis of the authors' estimation equation of the heat of vaporization for liquids with the residual function method, and with the liquid-free volume model as well as the theory of molecular dynamics. The results are compared with the recent methods proposed by Lei et al. and Klaas et al., and with the Jamieson equation and the modified Riedel equation.

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2. Derivation of the new model

Based on the absolute reaction rate theory of Eyring [13] and the free volume theory [14], Zhang and Liu [15] showed that the liquid viscosity is inversely proportional to the product of the probability P_E of containing activation energy E_a for a molecule and the probability P_V of possessing proper free volume around the molecule. On the basis of the ideas of Zhang et al. and the fact that the influences of temperature on thermal conductivity and viscosity have the same trend [16,17], referring to the thought of Teja and co-workers [9,18–20] that thermal conductivity and viscosity have analogous expressions, we conclude that thermal conductivity is inversely proportional to the product of P_E and P_V. Thus,

$$\lambda^{-1} = aP_E P_V \tag{1}$$

where P_E and P_V are expressed as Eyring's [13,21] and Ertl's [22] relations, respectively:

$$P_E = b\exp\left(-\frac{E_a}{RT}\right) \tag{2}$$

$$P_V = eV_f^n \tag{3}$$

According to [13,15], the molar activation energy E_a, and the liquid lattice energy E_c are in proportion as follows:

$$E_a = dE_c \tag{4}$$

By introducing Eqs. (2)–(4) into Eq. (1) and taking natural logarithm in Eq. (1), the following is obtained:

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$$\ln\lambda = -\ln(abe) - n\ln V_{\mathrm{f}} + \frac{\mathrm{d}E_{\mathrm{c}}}{RT} \tag{5}$$

Assuming that Eq. (5) is applicable to the vapor phase of a vapor–liquid equilibrium, and the vapor phase is dilute gases, then $E_{\mathrm{c}}^{\mathrm{V}} \to 0$, $V_{\mathrm{f}}^{\mathrm{V}} \to V$ [23]. Thus,

$$\ln\lambda^{\mathrm{V}} = -\ln(abe) - n\ln V \tag{6}$$

By subtracting Eq. (6) from Eq. (5), the following is obtained:

$$\ln\frac{\lambda}{\lambda^{\mathrm{V}}} = -n\ln\frac{V_{\mathrm{f}}}{V} + \frac{\mathrm{d}E_{\mathrm{c}}}{RT} \tag{7}$$

According to the theory of chemical engineering thermodynamics, chemical potentials are equal at vapor–liquid equilibrium and can be expressed by configuring partition function Q as follows [23]:

$$\left(\frac{\partial \ln Q^{\mathrm{L}}}{\partial N}\right)_{T,V} = \left(\frac{\partial \ln Q^{\mathrm{V}}}{\partial N}\right)_{T,V} \tag{8}$$

and

$$Q^{\mathrm{L}} = V_{\mathrm{f}}^{N}\exp\left(-\frac{E_{\mathrm{c}}}{RT}\right) \tag{9}$$

$$Q^{\mathrm{V}} = \frac{V^{N}}{N!} \tag{10}$$

Eqs. (9) and (10) are substituted into Eq. (8) and the Stirling approximation is adopted, with $V_{\mathrm{f}}=Nv_{\mathrm{f}}$ and $E_{\mathrm{c}}=N^2 f(V, T)$ [24]; the following relation is obtained:

$$\ln\frac{V_{\mathrm{f}}}{V} = \frac{2E_{\mathrm{c}}}{nkT} \tag{11}$$

Substituting Eq. (11) into Eq. (7), we get

$$\ln\frac{\lambda}{\lambda^{\mathrm{V}}} = \frac{BE_{\mathrm{c}}}{RT} \tag{12}$$

The liquid lattice energy E_{c} is approximately equal to the vaporization energy U_{V} [24] and the latter is related to the vaporization heat L_{V} as follows [1]:

$$E_{\mathrm{c}} \approx U_{\mathrm{V}} = L_{\mathrm{V}} - RT \tag{13}$$

According to the theory of molecular dynamics [1,2,4], the thermal conductivity of dilute gases is [11]

$$\lambda^{\mathrm{V}} = \frac{C(T/M)^{1/2}}{V_{\mathrm{c}}^{2/3}\Omega_{\mathrm{V}}} \tag{14}$$

where the collision integration Ω_{V} is expressed by the Neufeld–Janzen–Aziz equation [25] as follows:

$$\Omega_{\mathrm{V}} = 0.52487\exp(-0.97369T_{\mathrm{r}}) + 2.16178 \times \exp(-3.07001T_{\mathrm{r}}) + 1.1223T_{\mathrm{r}}^{-0.14874} \tag{15}$$

By taking the natural logarithm in Eq. (14) and defining

$$q \equiv \Omega_{\mathrm{V}}V_{\mathrm{c}}^{2/3}\left(\frac{T}{M}\right)^{-1/2} \tag{16}$$

the following is obtained:

$$\ln\lambda^{\mathrm{V}} = \ln C - \ln q \tag{17}$$

Introducing Eqs. (13) and (17) into Eq. (12) and arranging it, we get

$$\ln(\lambda q) = A + \frac{BL_{\mathrm{V}}}{RT} \tag{18}$$

We [26,27] have proposed the new equation to estimate the heat of vaporization for pure liquids based on the statistical thermodynamics theory as follows:

$$L_{\mathrm{V}} = \left(\frac{RT_{\mathrm{c}}SW}{P_{\mathrm{c}}}\right)\left(\frac{P_{\mathrm{c}}\ln P_{\mathrm{c}}}{\theta-1} - C^0 - \omega C'\right) \tag{19}$$

where

$$S = (10 + 3T_{\mathrm{r}} - 2T_{\mathrm{r}}^2)\left(\frac{\theta^2}{10\theta^2 + 3\theta - 2}\right)$$

$$W = \left(\frac{\theta - (T/T_{\mathrm{b}})}{\theta - 1}\right)^{0.38}$$

$$C^0 = 1.097\theta^{1.6} - 0.083$$

$$C' = 0.8944\theta^{4.2} - 0.139$$

$$\theta = \frac{T_{\mathrm{c}}}{T_{\mathrm{b}}}$$

$$T_{\mathrm{r}} = \frac{T}{T_{\mathrm{c}}}$$

The acentric factor in Eq. (19) is calculated by the Lee–Kesler equation [28].

Combining Eqs. (18) and (19), the new equation to calculate the thermal conductivity of pure liquids is obtained as follows:

$$\ln(\lambda q) = A + B\frac{SWT_{\mathrm{r}}^{-0.5}}{P_{\mathrm{c}}}\left(\frac{P_{\mathrm{c}}\ln P_{\mathrm{c}}}{\theta-1} - C^0 - \omega C'\right) \tag{20}$$

3. Results and discussion

By use of the least squares method, the two characteristic parameters A and B of a liquid in Eq. (20) can be determined from experimental data over the investigated temperature range. The values of A and B for some liquids are given in Table 1.

With the parameters A and B, we can use the most fundamental physical property data comprising T_{b}, T_{c}, P_{c} and V_{c} to calculate the thermal conductivity of some substances at any temperature according to Eq. (20). The thermal conductivity of 68 pure liquids including paraffins, olefins, alkynes, cycloparaffins, aromatics, alcohols, phenol, ethers, aldehydes, ketones, esters, organic acids, organic halides, organic nitrogen compounds and inorganic compounds were calculated from Eq. (20) over wide ranges of temperature,

Table 1
Calculated results of the thermal conductivity for some liquids in new model and comparison with other models

Liquids	Temperature range, T_r	No. of points	A	$B\times10$	ARD (%)					Refs.
					New model	Jamieson	Riedel	Klaas et al.	Lei et al.	
Methane	0.52034–0.97087	13	1.4089	2.7928	1.81	4.54	3.25	5.47	5.39	[17,29]
Ethane	0.60400–0.98200	10	1.6127	2.4383	2.22	5.29	2.41	4.86	6.62	[17]
Propane	0.48648–0.97192	18	1.7441	2.3323	1.67	4.46	3.36	5.68	7.10	[17,30]
Heptane	0.36653–0.96188	15	2.1913	1.9447	2.69	6.89	2.48	9.81	7.52	[17]
Dodecane	0.37696–0.64279	6	3.1570	1.2789	0.45	0.40	0.80	0.63	0.81	[17]
Octadecane	0.43376–0.76933	6	3.5220	1.0756	0.44	0.26	0.78	1.44	0.76	[17]
Isobutane	0.30176–1.0000	10	2.1895	3.6597	2.28	3.25	2.69	7.82	3.29	[17]
Ethylene	0.35379–0.99416	31	1.7212	2.4548	4.05	7.64	4.85	11.7	9.44	[16,17,29,30]
Propylene	0.61784–1.0000	10	1.9444	1.8065	2.44	2.74	3.83	8.13	4.57	[17]
1,2-Butadiene	0.64010–1.0000	10	2.1283	1.6715	2.81	1.51	1.26	8.44	4.43	[17]
Acetylene	0.62260–0.93940	9	0.8960	1.5149	2.80	1.82	2.11	1.01	1.27	[17]
Cyclopentane	0.38716–1.0000	16	2.2442	1.8584	3.15	2.71	2.23	8.90	9.49	[17,29]
Cyclohexane	0.51096–0.96544	26	1.8811	2.2775	1.84	3.82	4.27	3.76	6.37	[16,17,29]
Benzene	0.48551–0.76991	32	2.1291	2.0214	0.51	0.47	0.32	0.97	0.44	[16,29]
Toluene	0.32517–1.0000	55	2.3182	1.8082	1.13	2.31	1.71	5.96	5.11	[12,16,17,31]
Ethylbenzene	0.32110–0.99336	21	2.2851	1.812	3.47	3.56	3.92	7.42	9.29	[17,29]
Ethanol	0.33540–0.99370	47	2.2271	1.0833	2.15	2.57	2.62	3.97	4.53	[12,17,29,32]
1-Propanol	0.32253–0.99301	35	2.4982	1.0509	1.37	2.16	1.77	5.42	2.78	[17,29]
Isopropylalcohol	0.69907–0.99853	23	2.8081	0.5796	2.50	2.51	1.59	1.07	4.30	[17,29]
1-Butanol	0.35311–0.99599	27	2.4003	1.2127	1.67	4.21	1.15	7.39	4.01	[17,29,32]
1-Hexanol	0.40680–0.69369	6	2.9191	1.0876	0.56	0.71	0.72	0.49	1.26	[17]
1,2-Ethandiol	0.40310–0.77519	20	3.3144	0.4699	0.73	1.13	0.37	0.48	0.37	[17,29,36]
Phenol	0.65642–1.0000	10	2.8263	1.1278	0.78	4.29	3.79	7.57	1.90	[17]
Diethyl ether	0.37077–0.99143	25	2.3320	1.7930	2.72	3.58	3.39	7.25	7.02	[17,29]
Acetaldehyde	0.37560–0.63590	6	2.1557	1.6905	0.81	0.73	1.09	0.74	1.49	[17]
Acetone	0.38994–0.94460	22	2.0164	1.8298	2.92	3.73	1.62	6.38	2.69	[12,17]
Hexylic acid	0.41063–0.71129	6	2.7836	1.0551	0.47	0.50	0.77	0.57	0.70	[17]
Ethyl acetate	0.37869–1.0000	23	2.3842	1.6543	1.54	5.77	1.42	9.05	6.72	[12,17]
Dichloromethane	0.33951–0.73167	11	2.6737	1.4445	0.47	0.69	0.56	0.63	1.20	[29,33]
Chloroform	0.41601–0.94146	31	2.2981	1.8580	2.29	3.05	2.77	3.21	2.88	[12,17,29]
Carbon	0.45502–1.0000	48	2.0709	2.1419	1.27	3.34	1.08	7.30	4.27	[16,17,29]
tetrachloride	0.46450–0.68468	8	3.3051	0.7813	2.45	2.63	2.12	2.31	1.73	[29,33]
1,1,1-Trichloroethane										
trans-1,2-Dichloroethane	0.49347–0.72739	7	3.1732	0.4839	0.56	0.91	0.60	0.56	1.14	[33]
Refrigerant 12	0.46777–0.98753	43	1.8527	2.3514	2.80	5.24	2.80	6.92	7.28	[16,17,29,30]
Refrigerant 13	0.63418–1.0000	19	1.7917	2.5084	2.13	6.56	0.87	9.94	7.42	[16,17]
Refrigerant 14	0.51135–0.91202	4	1.8248	2.1823	1.54	3.62	2.38	5.66	4.07	[17]
Refrigerant 21	0.33939–0.97507	14	2.1534	1.8947	2.26	3.02	3.13	7.00	7.47	[17,29]
Refrigerant 22	0.46886–1.0000	26	1.8153	1.9471	3.26	5.89	3.06	11.8	9.28	[17,29,30]
Refrigerant 113	0.51950–0.74523	12	2.114	2.0101	0.35	0.62	0.27	0.88	0.27	[16]
Refrigerant 114	0.55658–0.89079	15	2.0464	2.1346	0.99	2.19	0.43	3.23	0.67	[16]
Refrigerant 142	0.47156–0.88660	18	1.8768	2.1910	1.77	2.39	1.00	4.17	1.38	[16]
Bromobenzene	0.40756–0.63138	14	2.3113	1.7919	0.59	0.53	0.80	0.99	2.55	[17,29]
Isopropyl amine	0.41628–0.61586	5	2.4879	1.4201	0.18	0.43	0.46	0.10	0.43	[17]
Aniline	0.39056–1.0000	13	2.7610	1.2666	1.60	6.01	4.90	9.10	1.74	[17,29]
Monoethanolamine	0.48502–0.72818	10	2.8984	0.5031	0.23	0.43	0.23	0.32	0.23	[31]
Nitrobenzene	0.41161–0.66435	16	2.7489	1.3467	0.94	0.77	1.19	1.69	8.46	[16,17,29]
Pyridine	0.39389–0.59230	5	2.1729	1.9853	0.27	0.43	0.36	0.60	0.56	[17]
Fluorine	0.59167–0.98958	10	1.5772	2.6547	1.96	6.21	1.62	9.44	3.42	[17]
Chlorine	0.57294–0.99724	10	1.7573	2.6278	1.26	11.7	0.74	10.2	3.96	[17]
Bromine	0.44521–0.85616	13	1.8202	2.2545	1.10	1.25	0.46	2.83	1.22	[17,33]
Helium	0.81118–0.96339	9	2.3282	−0.9993	0.27	8.35	0.17	0.29	0.97	[17]
Neon	0.56306–1.0000	15	1.3643	2.6846	3.99	8.14	4.85	9.99	3.47	[16,17]
Argon	0.57862–1.0000	29	1.2934	2.9397	1.19	5.91	1.56	9.82	4.47	[16,17,34]
Krypton	0.57307–0.95511	10	1.1234	3.0633	0.45	4.92	2.08	5.18	2.76	[16]
Xenon	0.55678–0.89748	11	1.1228	3.2282	0.80	2.23	1.52	3.54	1.71	[16,34]
Hydrogen	0.61330–1.0000	10	1.2783	2.3268	4.10	8.62	8.53	9.96	4.06	[17]
Oxygen	0.58151–0.96918	22	1.4937	2.7094	0.65	3.71	1.48	4.26	3.49	[16,17]
Air	0.45399–0.94268	35	1.5212	2.924	1.40	3.24	2.23	5.76	5.42	[17,33]
Carbon monoxide	0.54934–0.93872	10	1.3381	2.9623	0.43	3.69	3.02	4.66	3.45	[17]
Carbon dioxide	0.71190–1.0000	10	1.6437	1.9833	3.11	10.0	3.54	10.8	3.27	[17]

Table 1 (*Continued*)

Liquids	Temperature range, T_r	No. of points	A	$B\times 10$	ARD (%)					Refs.
					New model	Jamieson	Riedel	Klaas et al.	Lei et al.	
Sulfur dioxide	0.46425–0.92851	15	2.2273	1.8890	0.22	1.85	1.56	3.16	2.82	[16,17,36]
Hydrogen fluoride	0.63470–1.0000	9	1.7282	1.9170	1.23	3.79	1.62	3.62	3.29	[17]
Hydrogen chloride	0.53342–0.93962	10	2.001	2.5376	2.30	4.09	4.34	4.19	4.17	[17]
Hydrogen sulfide	0.57028–0.91116	9	1.0464	3.3193	2.07	3.94	2.67	5.52	4.92	[17]
Ammonia	0.59117–0.96166	38	1.9562	2.2783	1.40	3.86	1.95	3.59	2.69	[16,17,34]
Dowtherm J	0.34009–0.94491	9	3.4327	0.8740	1.17	1.54	0.33	0.25	0.55	[17]
Mercury	0.3574–0.59550	10	1.9630	1.3871	0.53	0.30	0.80	0.43	0.45	[17]
Water (0.1 MPa)	0.42198–0.57647	44	3.3653	0.4102	0.68	0.54	0.79	0.66	0.70	[29,33,35]
Water (5 MPa)	0.42198–0.57647	11	2.5115	0.4940	0.84	0.67	0.97	0.82	0.76	[33]
Water (20 MPa)	0.42198–0.57647	11	2.5063	0.5147	0.84	0.68	0.99	0.82	0.76	[33]
Total 68		1217			1.70	3.43	2.04	5.21	4.09	

even up to the critical temperature. The results are shown in Table 1. The reference data of thermal conductivity come from Vargaftik [16], Beaton [17], Jamieson [29], Lei et al. [12], Rohsenow [30], Digullio et al. [31] and Cai and others [32–36]. Table 1 also shows the comparison between the new model and other good models, such as the Jamieson equation [4–6], the modified Reidel equation [7] and the recent models proposed by Klaas et al. [10] and Lei et al. [11]. Comparing the calculated data and reference data of 68 pure liquids at 1217 temperature points, the overall average relative deviations of five models are, respectively, 1.70, 3.43, 2.04, 5.21, and 4.09%. The overall average relative deviation is calculated as follows:

$$\mathrm{ARD} = \left(\frac{100}{N_\mathrm{p}}\right)\sum\left[\frac{|\lambda_\mathrm{p}-\lambda_\mathrm{e}|}{\lambda_\mathrm{e}}\right] \quad (21)$$

Obviously, the new model is the best over the investigated temperature as well as substance range, and the modified Reidel equation takes the second place. The Jamieson equation is applicable to common liquids, but has notable errors when applied to lower paraffin hydrocarbons, some refrigerants, liquefied inert gas, diatomic gas and inorganic gas, especially when temperatures are close to the critical temperature. The model proposed by Klaas et al. can give good results when the temperature range is very limited. However, the calculated error rises sharply when the temperature range is extended, especially up to the critical temperature. Klaas et al., however, pointed out [10] that their model gives good results for the temperature range between the normal melting point and the normal boiling point of a substance. The method of Lei et al. has the same limitation as the model by Klaas et al.

4. Conclusions

Using our previous estimation equation of the heat of vaporization for liquids with the residual function method, and with the liquid free volume model as well as the theory of molecular dynamics, a new two-parameter model of thermal conductivity for liquids has been derived. With this new model, the thermal conductivity data for 68 kinds of liquids at 1217 temperature sets were tested. The results are compared with the recent models proposed by Lei et al., Klaas et al., and with the Jamieson equation and the modified Riedel equation. The comparison shows that the new model, which has a theoretical basis, is applicable to paraffins, olefins, alkynes, cycloparaffins, aromatics, alcohols, phenol, ethers, aldehydes, ketones, esters, organic acids, organic halides, organic nitrogen compounds, refrigerants, liquefied inert gas, inorganic compounds, and so on. The new model can give good results over a wide temperature range, even up to the critical temperature.

5. Nomenclature

a, b, d, e	constants
A, B, C	characteristic parameters of pure liquids
E_a	activation energy
E_c	lattice energy
k	Boltzmann constant
L_v	heat of vaporization
M	molecular weight
n	model parameter
N	molecular number
N_p	testing data sets
P	pressure
P_E, P_V	probability
Q	configuration partition function
R	gas constant
T	temperature
U_V	vaporization energy
V	volume
v_f	monomolecular free volume
V_f	mole free volume

Greek symbols

λ	thermal conductivity
Ω_V	collision integral
ω	acentric factor

Superscripts

L saturated liquid phase

V saturated vapor phase

Subscripts

b normal boiling point

c critical point

e experimental data

P calculated data

r reduced property

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