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An Empirical Correlation for Estimation of the Thermal Conductivity of Saturated Liquid Refrigerants

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Thermodynamic data on environmental refrigerants have attracted considerable interest in the optimization and design of heat compressors, exchangers and other refrigeration equipment. In this study, an optimization algorithm was used to obtain the constant parameters of the new empirical model by fitting them to the source databank. The model was used to calculate the constant coefficients of the new equation for 15474 data of 27 refrigerants (temperatures between 92 and 486 K) by fitting them with source data. The accuracy of the presented equation was compared with commonly used models. Results indicated that the developed model provides more accurate results than those of other considered equations, with an average absolute percentage deviation of 3.46%.

Keywords: Refrigerant, Correlation, Thermal conductivity, Empirical

INTRODUCTION

Transport properties of refrigerant fluids are essential in engineering design and application, especially in numerical calculations of heat transfer and fluid flow. Thus, the transport properties of working fluids, such as thermal conductivity, should be identified. However, the source data for thermal properties is small at present. Although many empirical correlations for calculating thermal conductivity are available, deviations of the database are typically unreasonably large to satisfy the requirements of engineering applications [1]. Thermal conductivity is fundamental for analyzing heat transfer in refrigerants. In particular, the amount of heat transfer coefficient increases with thermal conductivity [2].

Hence, many empirical correlations for the estimation of the thermal conductivity of refrigerants have been presented in the literature. These models include: 1) equations based on material properties, such as heat capacity and/or density and temperature [3,4] which are difficult to use for fluids with limited available data; 2) theoretical estimations investigating intermolecular distances [5]; and 3) correlations based on group contribution theory [6] or molecular descriptors [7-9], which are often unsuitable for all compounds [2].

Jaruskova and Kucerova [10] estimated several thermophysical properties, such as thermal conductivity using a nonlinear regression model with random parameters. Huber *et al.* [11] modified an extended empirical model for viscosity and thermal conductivity of 17 pure refrigerants originally developed by Ely and Hanley [12]. Latini *et al.* [13] developed a prediction method for tile thermal conductivity of halocarbon refrigerants in a saturated liquid state in the reduced temperature range from 0.3 to higher than 0.9. Latini and Sotte [14] compared three methods for estimating thermal conductivity in accordance with the database of the National Institute of Standards and Technology (NIST) Chemistry WebBook [15]; this database contains all common refrigerants belonging to the methane, ethane, and propane series. Amooey [16] developed a

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simple equation as a function of reduced temperature, molecular mass, diminished pressure, and acentric factor; this author applied this equation to predict the thermal conductivity of refrigerants over extensive temperature and pressure using 915 data. Nicola *et al.* [17] modified the Kardos equation, specifically for the saturation line of liquid and vapor states of refrigerants to estimate thermal conductivity.

In this work, we propose a simple substance-dependent equation based on liquid-vapor equilibrium databank. The new model accurately reproduces the thermal conductivity behavior over a wide range in the liquid-vapor region. On the basis of this model, an equation is also established. The thermal conductivity data used in this study were obtained from the National Institute of Standards and Technology Chemistry WebBook [15]. The new equation can estimate experimental data that are unused in regression analyses with low deviation. In contrast to the models in the literature, the proposed model is only a function of reduced temperature.

THERMAL CONDUCTIVITY CORRELATIONS

Some techniques for estimating thermal conductivity as a function of temperature, molecular weight and critical properties are available in literature. In this section, commonly used correlations for thermal conductivity estimation are presented.

Scheffy and Johnson Model

In 1961, Scheffy and Johnson in [18] presented a simple equation as a function of temperature, melting temperature and molecular weight as indicated in Eq. (1),

$$\lambda = 1.951\frac{1-0.00126(T-T_{fus})}{T_{fus}^{0.216}M^{0.3}} \quad (1)$$

where λ is thermal conductivity in W m^{-1} K^{-1}, T is the temperature in Kelvin, T_{fus} is the temperature of fusion in Kelvin, and M is molecular weight in g mol^{-1}.

Latini-Sotte Model

In 2011, Latini and Sotte [19] presented a developed model to show the relationship between thermal conductivity and the golden ratio (Eq. (2)),

$$\lambda = A\left[\sqrt{5}\frac{(\Phi-T_r)^2}{\Phi+T_r}\right]^a \quad (2)$$

where Φ is the golden ratio, the well-known value linked with Fibonacci's sequence expressed as follows:

$$\Phi = \frac{1+\sqrt{5}}{2} = 1.618033... \quad (3)$$

"*a*" assumes a unique value for each organic family (in the case of refrigerants, 0.60, 0.66 and 0.55 for fluids of the methane, ethane, and propane series, respectively). *"A"* represents the value of the thermal conductivity of fluids at the reduced temperature (*Tr*). This value corresponds to the mantissa of the golden ratio:

$$A = \lambda(T_r = \Phi - 1) = \lambda(T_r \cong 0.618033...) \quad (4)$$

Sato-Riedel Model

Sato-Riedel model [8] is a simple equation that requires temperature, normal boiling point temperature, molecular mass, and critical temperature. The Sato-Riedel model is expressed in Eq. (5),

$$\lambda = \frac{1.1053}{M^{1/2}}\frac{3+20(1-T_r)^{2/3}}{3+20(1-T_{br})^{2/3}} \quad (5)$$

where T_{br} is the reduced boiling temperature.

Gharagheizi *et al.* Model

To accurately estimate the thermal conductivity and to cover a significantly wider number of chemical families, an equation has been recently proposed by Gharagheizi *et al.* [20] adopting a mathematical strategy:

$$\lambda = 10^{-4}\left[10\omega + 2P_c - 2T + 4 + 1.908\left(T_b + \frac{1.009.B^2}{M^2}\right) + \frac{3.9287.M^4}{B^4} + \frac{A}{B^8}\right] \quad (6)$$

where

$$A = 3.8588\ M^8\ (1.0045B + 6.5152M - 8.9756) \quad (7)$$

and

$$B = 16.0407M + 2T_b - 27.9074 \quad (8)$$

This model is also applicable to estimate thermal conductivity of amines, silanes/siloxanes, inorganic compounds, sulfides/thiophenes, mercaptans, epoxides, peroxides, nitriles, elements, and aldehydes.

Sastri and Rao Model

Sastri and Rao [6] proposed an equation on the basis of the group contribution theory to calculate the thermal conductivity at the normal boiling point, λ_b (Eq. (9)),

$$\lambda = \lambda_b \, b^{1-\left(\frac{1-T_r}{1-T_{br}}\right)^n} \quad (9)$$

where b and n are assumed to be 0.16 and 0.2, respectively, except for alcohols and phenols.

NEW CORRELATIVE MODEL FOR THERMAL CONDUCTIVITY

A wide database of thermal conductivity is available in literature [15] because many organic and inorganic compounds have been used for many years.

This study aimed to determine a new simple equation for the thermal conductivity of liquid refrigerants. A four-constant nonlinear equation with high precision in the estimation of the vapor-liquid equilibrium data was developed. After multiple analyses of regression, an empirical correlation was suggested in Eq. (10),

$$\lambda = a + b.T_r^{0.0618} + c.T_r + d.T_r^{1.0618} \quad (10)$$

In Eq. (10), T_r is a dimensionless reduced temperature and λ is the thermal conductivity in W m^{-1} K^{-1}. The coefficients a, b, c and d are constant parameters obtained using the Levenberg-Marquardt algorithm (most accurate algorithm of Back-Propagation method available in many fitting software programs) which minimize the sum of the squared differences between the observed and predicted values of dependent variables [21].

In the first step, after considering thermophysical relations between thermal conductivity and reduced temperature in each refrigerant, a preliminary equation is achieved through trial and error. After investigating the standard deviations of each coefficient, we determined that the equation terms are changed. The last accurately tuned coefficients with the minimal deviation are listed in Table 1.

The new empirical model is much simpler with fewer terms than Gharagheizi *et al.'s* model. The Scheffy-Johnson, Sato-Riedel, and Gharagheizi *et al.* models used molecular weight and boiling temperature, whereas the new empirical model adopted only reduced temperature to estimate the thermal conductivity of refrigerants.

RESULTS AND DISCUSSION

Results of New Model

We carried out calculations for 27 pure refrigerants. The values of thermal conductivity, molecular weight, critical temperature and acentric factor (for calculation of literature models) were taken from the databank (NIST Chemistry Webbook) [15].

To verify the high accuracy of the new empirical model, the calculated thermal conductivity values versus the corresponding values in NIST [15] has been presented in Fig. 1.

Figure 1 indicates that most of data points are nearly diagonal, and has low deviation from the values in the NIST databank.

To exhibit the accuracy of literature correlations, the predicted thermal conductivity values obtained from literature correlations versus values obtained from the NIST databank [15] are plotted in Figs. 2-6.

The average absolute relative deviation (AARD%) of the thermal conductivity obtained by the newly proposed equation and literature models against the NIST values are presented in Table 2.

The newly proposed equation is more accurate than those of other literature models for almost all types of refrigerants considered in this work.

To further compare, the accuracy of four literature models with the new correlation, Fig. 7 presents the average absolute relative deviation (AARD%), Fig. 8 presents the average relative deviation (ARD%), Fig. 9 presents the

Table 1. Critical Properties, Temperature Range and Tuned Coefficients of the New Proposed Model

Type	No. of data	T_{min} (K)	T_{max} (K)	*a*	Dev.% of *a*	*b*	Dev.% of *b*	*c*	Dev.% of *c*	*d*	Dev.% of *d*
R11	531	198.1	470.6	1.3214	0.21	-1.2977	0.03	0.9384	0.18	-0.9216	0.02
R12	500	160.9	384.6	1.4823	0.14	-1.4560	0.09	0.6876	0.12	-0.6754	0.01
R13	600	92	301.6	1.0058	0.71	-0.9958	0.01	2.2854	0.61	-2.2626	0.01
R14	599	98.9	227.1	1.8527	0.33	-1.8232	0.01	0.6158	0.21	-0.606	0.02
R22	499	158.4	368.8	1.6335	0.10	-1.6069	0.03	1.4977	0.08	-1.4733	0.00
R23	200	118	298.9	40.5413	0.17	-46.405	0.05	60.738	0.13	-54.829	0.09
R32	600	136.3	350.9	1.7899	0.34	-1.7683	0.03	4.6771	0.30	-4.6207	0.01
R41	600	175	317	3.7661	0.21	-3.7190	0.08	3.5677	0.17	-3.5231	0.03
R113	599	236.9	486.7	1.0738	0.12	-1.0445	0.00	0.2198	0.15	-0.2138	0.02
R114	600	273.1	418.6	1.0365	0.44	-1.0101	0.02	0.4169	0.52	-0.4063	0.01
R115	597	173.7	353.1	1.3126	0.52	-1.2815	0.03	0.2657	0.41	-0.2594	0.01
R116	583	175.7	292.6	1.0152	0.72	-0.9993	0.09	1.3152	0.60	-1.2946	0.00
R123	600	166	456.3	1.1224	0.71	-1.1037	0.08	1.0893	0.65	-1.0711	0.01
R124	600	120	394.9	0.9098	0.15	-0.8998	0.09	2.1208	0.10	-2.0974	0.00
R125	600	172.5	338.9	1.5336	0.09	-1.5071	0.02	0.9704	0.10	-0.9536	0.02
R134a	600	169.8	373.8	1.8522	0.23	-1.8172	0.03	0.7932	0.21	-0.7782	0.01
R141b	600	169.6	476.9	1.1651	0.45	-1.1405	0.04	1.1064	0.59	-1.083	0.03
R142b	600	142.7	409.8	1.2403	0.62	-1.2225	0.01	1.6949	0.55	-1.6706	0.00
R143a	600	161.3	345.5	1.8279	0.03	-1.7820	0.02	0.2987	0.07	-0.2912	0.00
R152a	600	154.5	386	2.0634	0.19	-2.0093	0.01	0.3665	0.18	-0.3569	0.01
R218	600	125.4	344.6	0.5322	0.32	-0.5254	0.08	1.7789	0.28	-1.756	0.03
R227ea	600	146.3	375.6	0.6055	0.12	-0.5956	0.01	1.7627	0.17	-1.7339	0.00
R236ea	601	242	412.4	1.8354	0.91	-1.7915	0.12	3.76×10^{-5}	1.11	3.67×10^{-5}	0.04
R236fa	584	185.3	397.7	1.3635	1.03	-1.3300	0.15	0.5084	1.21	-0.4959	0.04
R245ca	581	205.3	444.7	1.4237	0.60	-1.3831	0.04	0.611	0.51	-0.5936	0.02
R245fa	600	200	426.8	1.4202	0.13	-1.3886	0.01	0.906	0.080	-0.8858	0.00
RC318	600	233.3	388.1	0.6624	0.42	-0.6506	0.03	1.6352	0.36	-1.6059	0.01

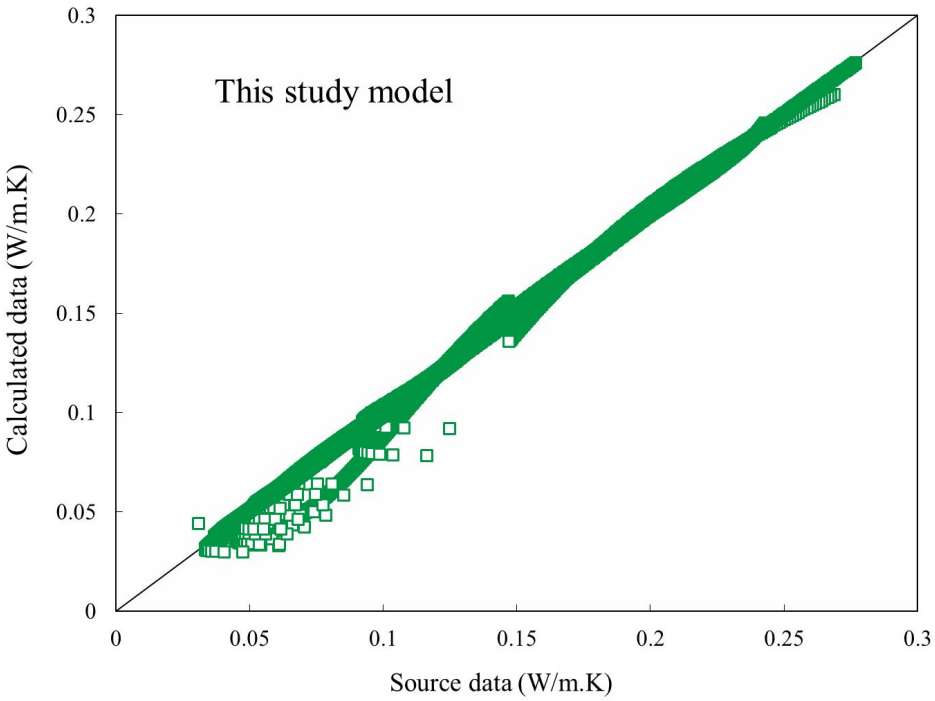

Fig. 1. Accuracy of this study model *versus* values from databank.

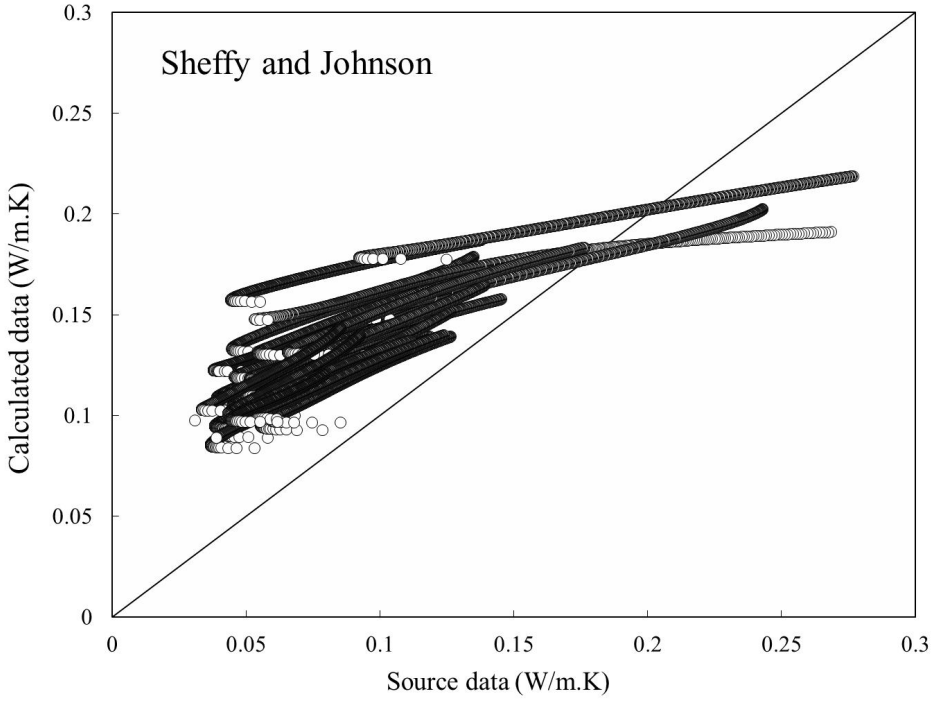

Fig. 2. Accuracy of Sheffy and Johnson model *versus* values from databank.

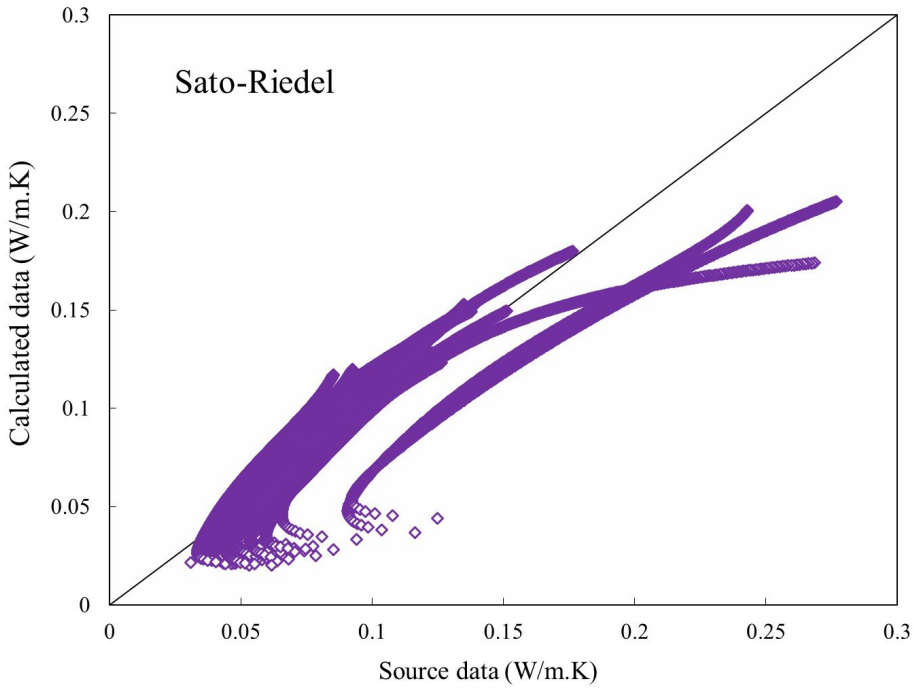

Fig. 3. Accuracy of Sato-Riedel model *versus* values from databank.

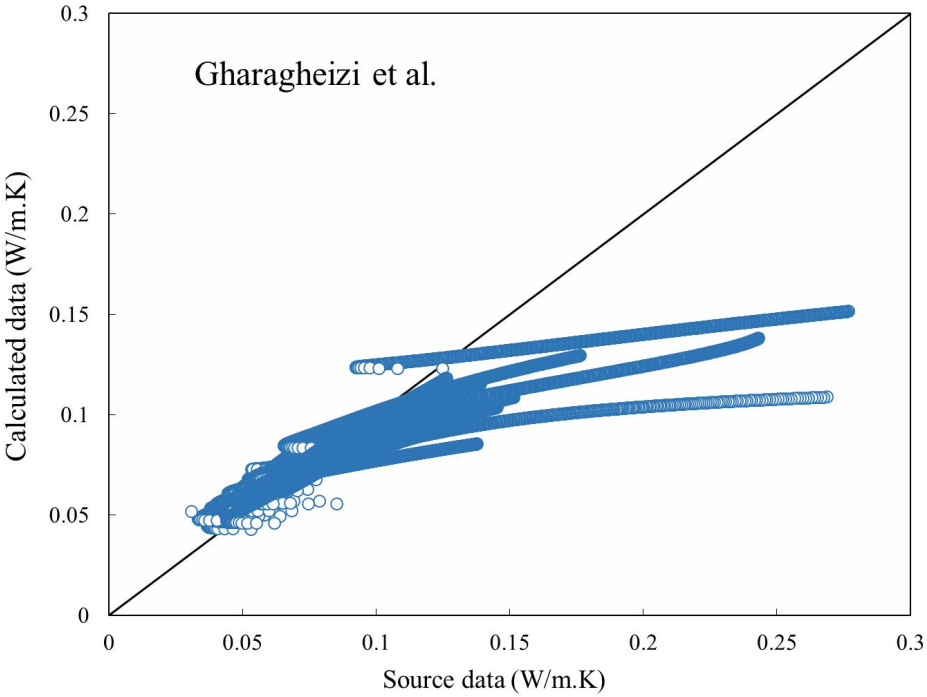

Fig. 4. Accuracy of Gharagheizi *et al.* model *versus* values from databank.

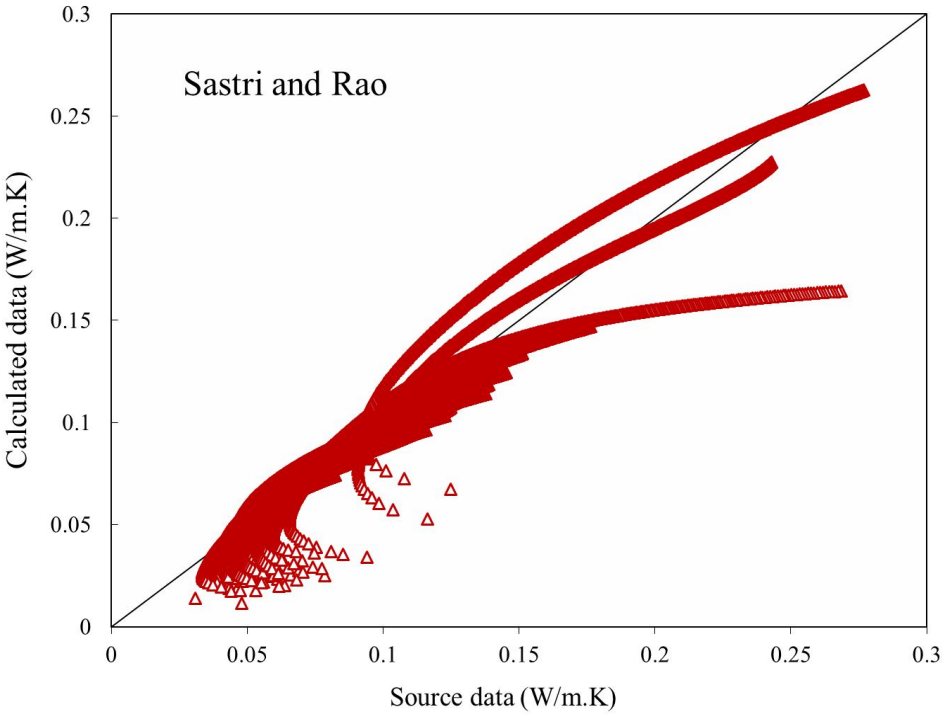

Fig. 5. Accuracy of Sastri-Rao model *versus* values from databank.

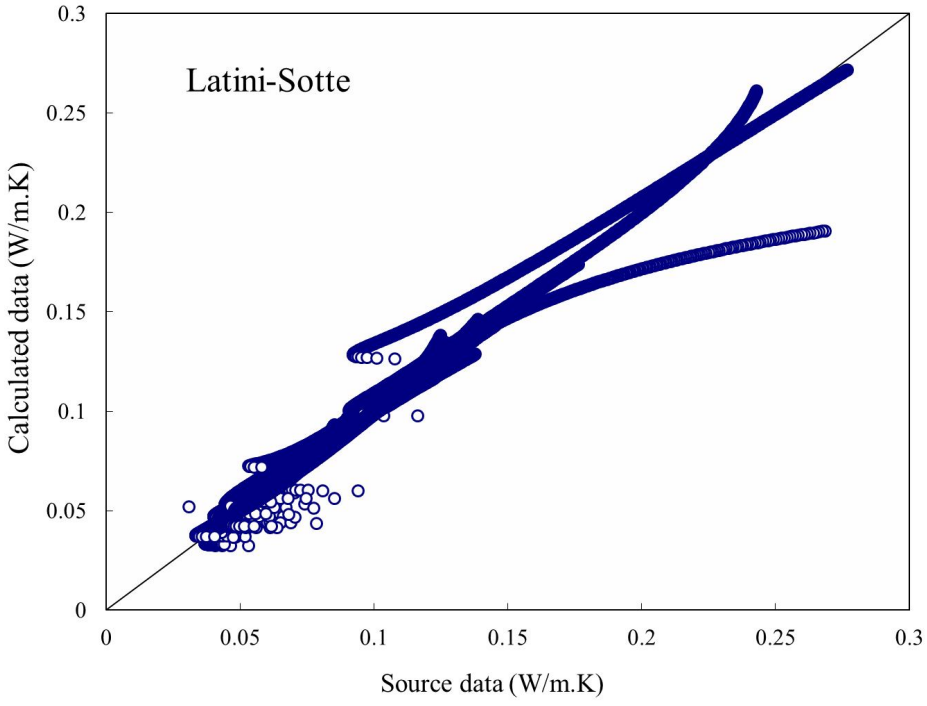

Fig. 6. Accuracy of Latini-Sotte model *versus* values from databank.

Table 2. AARD% of Literature Equations Compared to the New Model

No.	Substance	Sato-Riedel	Gharagheizi *et al.*	Sastri-Riedel	Latini-Sotte	This study
1	R11	7.5	8.02	6.72	2.81	0.59
2	R12	13.19	13.4	8.47	5	0.64
3	R13	17.32	17.27	9.53	4.19	3.02
4	R14	20.74	21.15	11.27	8.48	1.10
5	R22	5.35	15.49	6.62	3.18	0.63
6	R23	10.24	27.35	11.04	9.03	2.09
7	R32	21.48	27.41	6.04	3.2	1.61
8	R41	24.02	25.54	11.71	9.92	0.63
9	R113	20.97	20.6	4.91	2.75	0.55
10	R114	19.79	19.8	4.94	6.28	0.77
11	R115	19.96	12.55	5.93	0.63	1.27
12	R116	18.62	12.86	4.08	1.63	0.42
13	R123	13.5	9.68	6.92	1.18	1.44
14	R124	13.63	11.06	8.22	2.13	3.16
15	R125	9.95	12.77	6.34	1.97	0.70
16	R134a	5.86	14.55	6.73	1.65	0.57
17	R141b	13.65	6.02	6.76	4.82	1.90
18	R142b	14.21	12.26	7.73	1.08	1.89
19	R143a	15.03	12.61	5.96	0.97	0.93
20	R152a	10.61	13.83	6.6	1.68	0.47
21	R218	27.1	16.09	4.25	3.71	1.24
22	R227ea	19.31	6.65	3.6	2.32	1.24
23	R236ea	6.93	8.22	5.89	8.03	0.33
24	R236fa	10.34	7.11	5.37	2.17	0.64
25	R245ca	8.99	10.48	4.64	1.27	0.49
26	R245fa	7.18	8.75	4.6	1.4	0.41
27	RC318	10.22	3.72	3.89	2.17	1.11

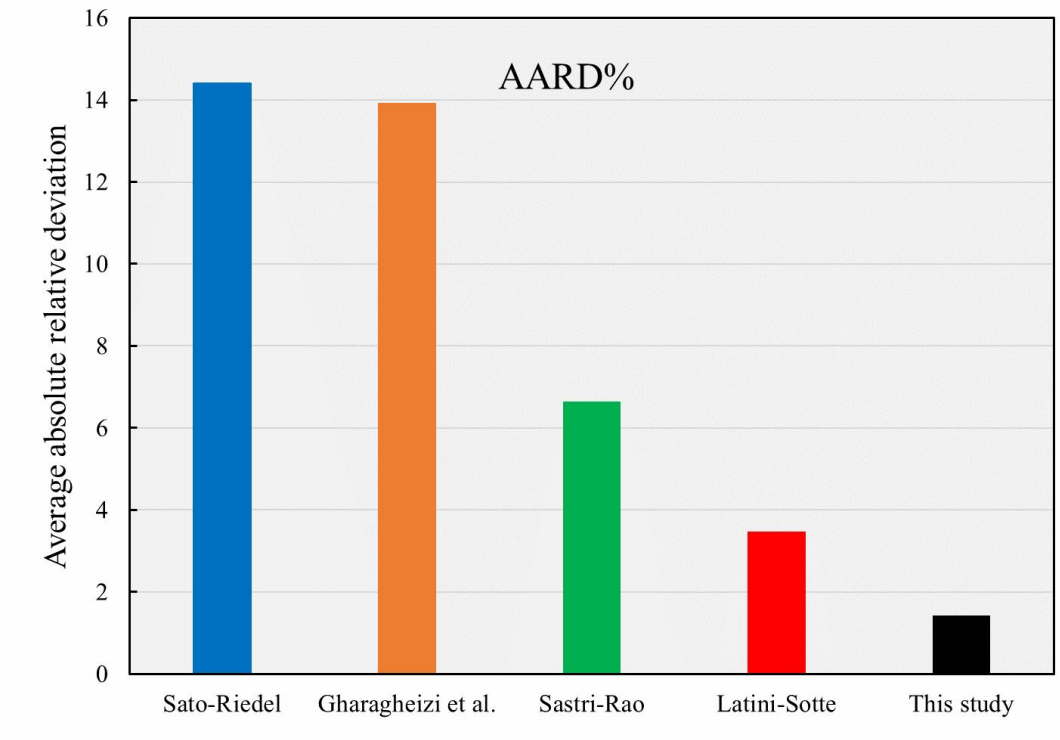

Fig. 7. AARD% of this study compared with those of other methods.

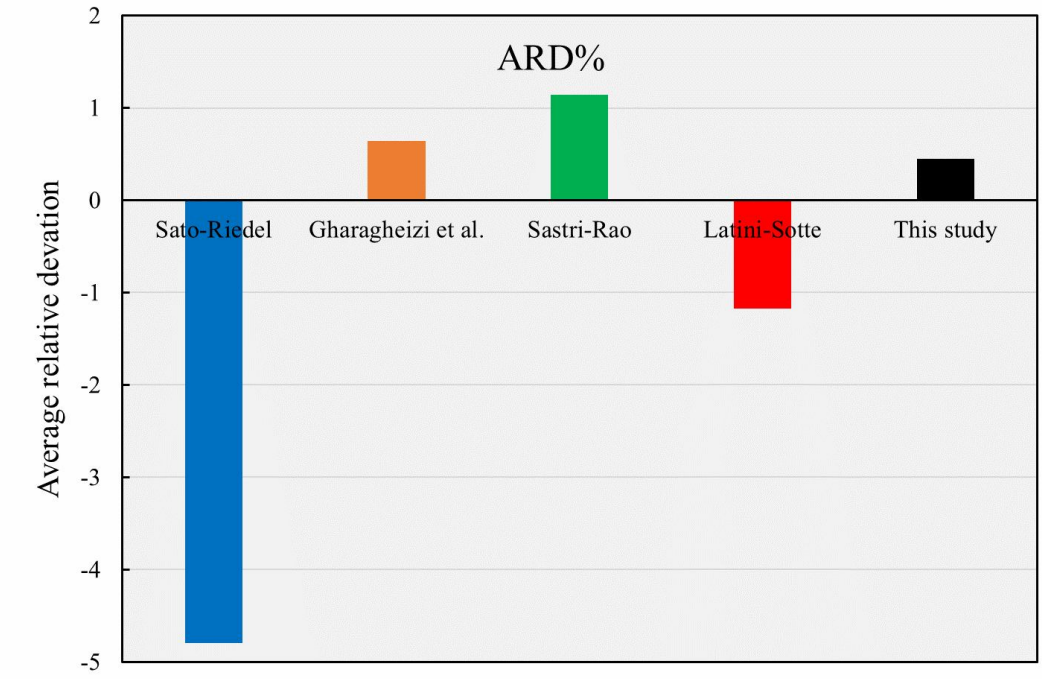

Fig. 8. ARD% of this study compared with those of other methods.

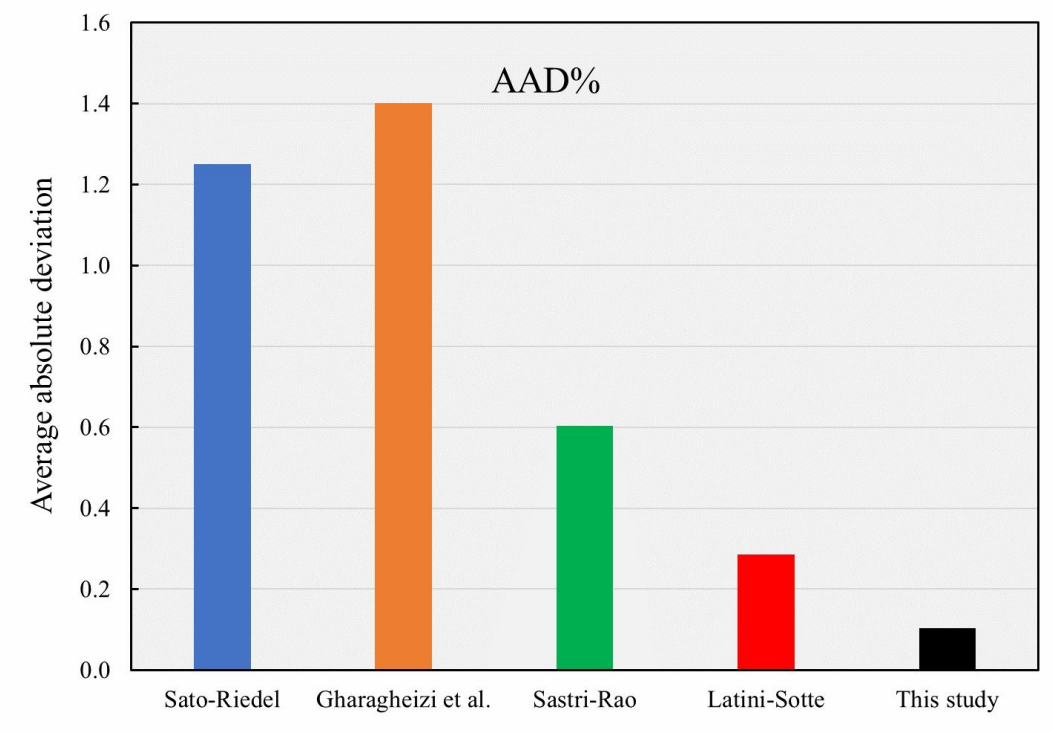

Fig. 9. AAD% of this study compared with those of other methods.

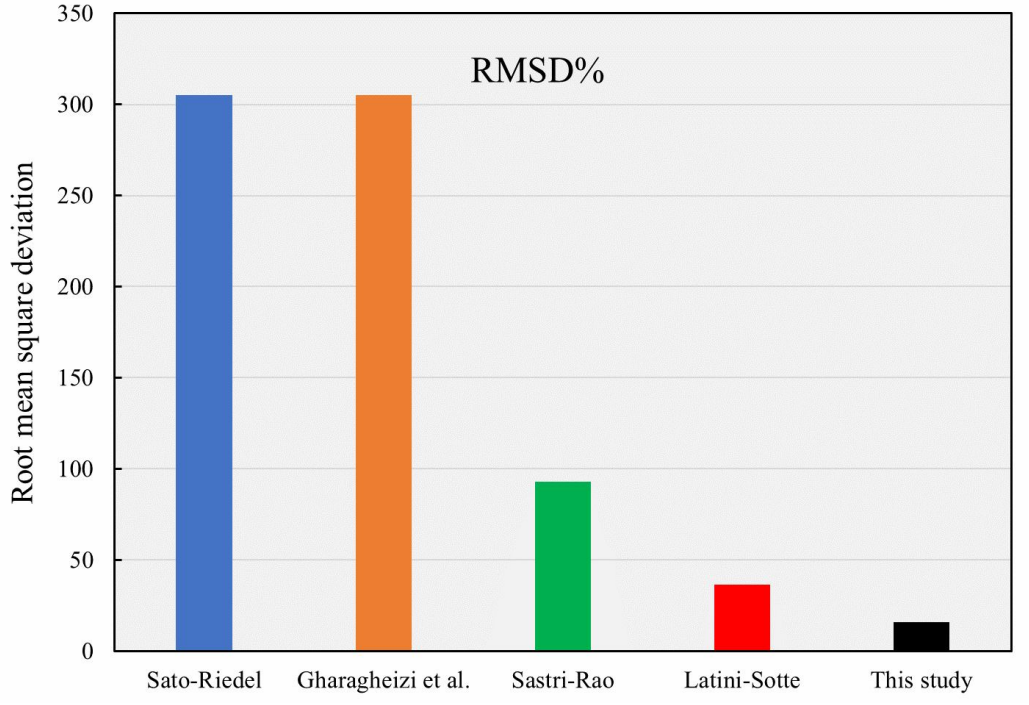

Fig. 10. RMSD% of this study compared with those of other methods.

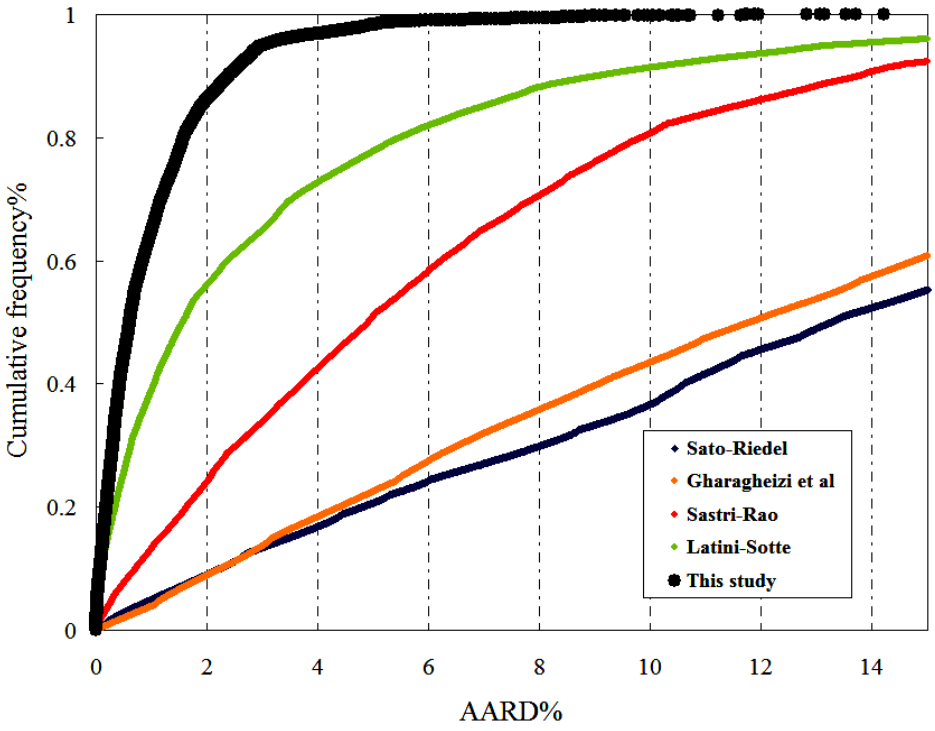

Fig. 11. Cumulative frequencies of various models in calculating thermal conductivity as a function of AARD%.

average absolute deviation (AAD%), and Fig. 10 presents the root mean square deviation (RMSD%) of models for the refrigerants under study. The criteria for deviations are calculated as shown in Eqs. (11)-(14):

$$AARD\% = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{\lambda_{r,i,\exp}-\lambda_{r,i,calc}}{\lambda_{r,i,\exp}}\right|\times 100 \tag{11}$$

$$ARD\% = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{\lambda_{r,i,\exp}-\lambda_{r,i,calc}}{\lambda_{r,i,\exp}}\right)\times 100 \tag{12}$$

$$AAD\% = \frac{1}{N}\sum_{i=1}^{N}\left|\lambda_{r,i,\exp}-\lambda_{r,i,calc}\right|\times 100 \tag{13}$$

$$RMSD = \frac{1}{N}\sqrt{\sum_{i=1}^{N}\left(\frac{\lambda_{r,i,\exp}-\lambda_{r,i,calc}}{\lambda_{r,i,\exp}}\right)^{2}}\times 100 \tag{14}$$

As shown in Figs. 7-10, the developed equation is more accurate than those of four other literature models in the estimation of the thermal conductivity.

Figure 11 shows the cumulative frequencies of the proposed and literature models versus average absolute relative deviations.

The new model successfully predicts 86.9% and 97% of all experimental data with an AARD% of less than 2 and less than 4, respectively. Only 0.001% of the thermal conductivity data are predicted with AARD% of more than 10% by the new model. Latini-Sotte model, the second most accurate model, predicts 56% and 73% of the data with an AARD% of less than 2 and 4, respectively. Finally, the superiority of this new model over the other literature models was verified for almost all refrigerants.

Applicability of New Model

The experimental data [22-25] of the four refrigerants include R32, R124, R125, R141b, R123, R134a, R22, R142b and R152a. These data are unused in the regression analysis of the proposed correlation, nonetheless, to estimate the applicability of the new correlation to calculate the thermal conductivity of various refrigerants, these data are used for calculating the thermal conductivity to validate

Table 3. Properties of Validation Data Set of Nine Refrigerants [22-25]

Authors (Year)	Refrigerant	T range (K)	λ range (W m^{-1} K^{-1})	No of data	AARD%	Ref.
Assael *et al.* (1995)	R32	252.6-312.8	0.170-0.111	27	3.124	[22]
	R124	252.9-332.9	0.058-0.091	35	4.427	
	R125	253.0-313.5	0.058-0.079	20	4.872	
	R141b	253.2-313.3	0.088-0.108	27	3.843	
Assael *et al.* (1993)	R22	253.2-333.2	0.067-0.104	5	1.504	[23]
	R123	253.2-333.2	0.068-0.090	5	1.276	
	R134a	253.2-333.2	0.066-0.101	5	0.646	
Kim *et al.* (1993)	R22	223.2-323.2	0.071-0.118	5	0.186	[24]
	R142b	223.2-323.2	0.074-0.110	5	1.754	
	R152a	223.2-323.2	0.087-0.133	5	3.721	
Tsvetkav *et al*. (1994)	R134a	169.9-290.1	0.084-0.145	19	1.167	[25]
	R123	159.3-291.3	0.079-0.118	19	0.887	
	R125	172.7-290.0	0.063-0.116	16	3.524	

the new equation.

The temperature range, thermal conductivity range, number of data points, and AARD of the aforementioned refrigerants are summarized in Table 3. Table 3 verifies that the new equation has a low deviation over new experimental data [22-25], which are unused in the regression analysis with an AARD of 3.01%.

Given the weak dependency of liquid-phase properties to the pressure changes, a new equation (Eq. (10)) with the same tuned coefficients, displayed in Table 1, is evaluated using a new set of 27304 data from the databank [15] for a compressed liquid phase under a saturation temperature. Table 4 lists the accuracy of the proposed model over the compressed liquid-state data. The source of 27304 data and calculation of the new model are presented in "Supplementary File."

CONCLUSIONS

Most commonly used thermal conductivity prediction models for liquid refrigerants were evaluated. Undesirable estimation deviations were obtained using the Sato-Riedel and Gharagheizi *et al.* equations over a wide range of temperature. The Latini-Sotte model generally provides good prediction accuracy relative to the other models. Results indicated that the newly proposed model is the best model for the 27 pure liquid refrigerants. The proposed correlation presents more number of constants than those of other literature models, however, results indicated the superiority of this new model over all other models in terms of calculating the thermal conductivity of pure liquid refrigerants with an AARD of 0.6%. The new model was evaluated using the new 193 experimental data

of the nine refrigerants. The new data were unused in the regression analysis with an AARD of 3.01%. Then, this model was evaluated using more than 27000 data of compressed liquid-state refrigerants under a saturation temperature with an AARD of 0.6%.

Nomenclature

λ	Thermal conductivity
λ_{exp}	Experimental thermal conductivity
λ_{calc}	Calculated thermal conductivity
T	Temperature
T_{fus}	Temperature of fusion
T_r	Reduced temperature (T/T_c)
T_{br}	Reduced boiling temperature (T_b/T_c)
T_b	Boiling temperature
T_c	Critical temperature
P_c	Critical pressure
M	Molecular weight
N	Number of data points
Φ	Golden ratio
ω	Acentric factor

Abbreviations

AARD	Average absolute relative deviation
AAD	Average absolute deviation
ARD	Average relative deviation
RMSD	Root mean square deviation

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