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Correlations for Liquid Thermal Conductivity of Low GWP Refrigerants in the Reduced Temperature Range 0.4 to 0.9 from Saturation Line to 70 MPa

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Abstract

This paper presents a literature survey of the experimental thermal conductivity data for environmentally friendly refrigerants, namely HydroFluoroOlefins (HFOs) and HydroChloroFluoroOlefins (HCFOs), in the liquid phase. A total of 2073 experimental data for six alternative refrigerants, i.e. R1233zd(E), R1234yf, R1234ze(E), R1234ze(Z), R1224yd(Z), and R1336mzz(Z), was collected in the temperature range from 203.18 K to 434.99 K and in the pressure range from 0.10 MPa to 66.62 MPa. Literature correlations not considering the pressure dependence were compared for reduced pressures lower than 1. Di Nicola et al. (2014a) correlation was re-fitted and new coefficients expressly dedicated to low global warming potential refrigerants were given. The proposed equation gives an overall deviation of 1.78 % for the data measured at reduced pressure up to 1. The liquid thermal conductivity dependence on pressure was also considered adding new terms on Di Nicola et al. (2014a) correlation and on Latini and Sotte (2012) correlation. The proposed equations give overall deviation of 1.45 % and 1.99 %, respectively, for the complete dataset.

Keywords: corresponding states principle; hydrofluoroolefin; global warming potential; transport properties.

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Nomenclature

Latin symbols

a	Coefficient of Latini and Sotte (2012) correlation and Di Nicola et al. (2014a) correlation
A	Parameter of Latini and Sotte (2012) correlation and Gharagheizi et al. (2013) correlation
A_0	Parameter of Latini and Sotte (2012) correlation
A_1	Parameter of the pressure dependent correlation proposed by Latini and his coworkers
A'	Coefficient of Latini and Sotte (2012) correlation
b	Coefficient of Di Nicola et al. (2014a) correlation
B	Parameter of Gharagheizi et al. (2013) correlation
B_i	Coefficients of Amooey (2017) correlation
c	Coefficient of Di Nicola et al. (2014a) correlation
d	Coefficient of Di Nicola et al. (2014a) correlation
e	Coefficient of Di Nicola et al. (2014a) correlation
f	Coefficient of the proposed pressure dependent correlation
f_0	Coefficient of the proposed pressure dependent correlation
g	Coefficient of the proposed pressure dependent correlation
M	Molar mass (kg mol^{-1})
N	Number of experimental points
P	Pressure (MPa)
Q	Coefficient of Missenard (1970) correlation
T	Temperature (K)
T_b	Normal boiling point temperature (K)

Greek Symbols

α	Coefficient of Latini and Sotte (2012) correlation
α_1	Coefficient of the pressure dependent correlation proposed by Latini and his coworkers
β	Coefficient of Latini and Sotte (2012) correlation
β_1	Coefficient of the pressure dependent correlation proposed by Latini and his coworkers
γ	Coefficient of Latini and Sotte (2012) correlation
λ	Thermal conductivity (W m^{-1} K^{-1})
λ_0	Coefficient of Di Nicola et al. (2014a) correlation
ω	Acentric factor
μ	Dipole moment (D)
Φ	Golden ratio

Subscripts/Superscripts

c	Critical
calc	Calculated
exp	Experimental
r	Reduced

Acronyms

AARD	Average Absolute Relative Deviations
CSP	Corresponding states principle
EU	European Union

GEM	Gene expression programming
GHG	Greenhouse gas
GWP	Global warming potential
HFC	Hydrofluorocarbons
HCFO	Hydrochlorofluoroolefins
HFO	Hydrofluoroolefins
MARD	Maximum absolute relative deviations

1. Introduction

In the last decades, the refrigeration industry focused on the research of low Global Warming Potential (GWP) replacement refrigerants for HydroFluoroCarbons (HFCs), with the aim of reducing the environmental impact caused by their emissions. In fact, different environmental regulations have been proposed to decrease the emissions of fluorinated GreenHouse Gases (GHGs), such as HFCs, in the coming years. At the European Union (EU) level, the Regulation (EU) No. 517/2014 (2014) (F-gas Regulation) imposed the GWP limit at 150 for the refrigerants used in several refrigeration and air conditioning vapor compression systems, aiming to decrease at least two thirds of the 2010 emissions of fluorinated GHGs by 2030. More recently, a proposal to ratify the Kigali amendment to the Montreal Protocol (UNEP, 2016), to phase-down HFCs production and use, was adopted by the European Commission (European Commission, 2017). A comprehensive search of potential low GWP synthetic refrigerant alternatives for air conditioning, heat pumping, and refrigeration equipment was carried out by NIST researchers (Domanski et al., 2017; McLinden et al., 2017, 2014). The authors identified only a limited number of low GWP fluids showing the suitable combination of environmental, safety, and thermodynamic characteristics. Many of the selected fluids are halogenated alkenes, namely HydroFluoroOlefins (HFOs) and HydroChloroFluoroOlefins (HCFOs). However, since none of the selected alternative refrigerants is ideal in all respects and most of them are flammable, it is necessary a trade-off between the different characteristics. Consequently, the interest in blends containing conventional and alternative refrigerants has increased (Bell et al., 2019).

Thermal conductivity, λ, of fluids is an important transport property for several phenomena of scientific and industrial interest. For refrigerants, its knowledge is fundamental for modelling boiling and condensation heat transfer processes. The experimental data of this property for many liquids and gasses are not available or are not sufficiently reliable. In this regard, Bobbo et al. (2018) showed that only a limited number of experimentally-determined thermal conductivity values for low GWP

refrigerants and their blends is available in the scientific literature. Consequently, estimation methods are necessary to describe the thermal conductivity of such fluids.

During the last century, different theoretical models to predict the thermal conductivity of liquids, which take into account intermolecular distances (Bridgman, 1923; Kardos, 1934) or the degree of association of fluids (Vargaftik, 1994), have been developed. However, unlike the ones commonly used for the gaseous state (Chapman, 1912; Tsederberg and Cess, 1965), the theory-based formulations to describe the liquid thermal conductivities are usually of poor practical use and inaccurate (Poling et al., 2001). In fact, these theoretical expressions for the liquid state can involve excessive mathematical complication or can provide large errors. For these reasons, several empirical and semi-empirical models to determine the liquid thermal conductivity have been developed for chemical and engineering purposes using different approaches, such as correlations based on group contribution or Corresponding States Principle (CSP) (Assael et al., 1989; Baroncini et al., 1979; Hopp and Gross, 2019; Latini and Pacetti, 1977; Lazzús, 2015; Lv et al., 2020; Poling et al., 2001). In particular, some estimation models were specifically developed for the description of the thermal conductivity of liquid refrigerants (Amooey, 2017; Assael et al., 2000; Di Nicola et al., 2018, 2014a; Huber et al., 2003; Khosharay et al., 2018; Latini and Sotte, 2012; Yang et al., 2020).

The two properties that mostly affect the liquid thermal conductivity are temperature and pressure. In general, thermal conductivity of liquids decreases with increasing temperature, although for some substances, such as water or other aqueous compounds, this trend does not occur. The variation with temperature is generally linear from the normal melting point to the normal boiling point and over. Instead, there is a different behavior near the critical point, which represents a singularity. It has to point out that empirical models developed for the liquid thermal conductivity are generally not able to predict the different behavior near the critical point, thus they have been successfully applied to temperatures lower than the critical one (Poling et al., 2001). Unlike temperature, thermal conductivity increases with pressure, even if this dependence is usually negligible up to 5 - 6 MPa

(Poling et al., 2001). Other models available in literature also require additional fluid properties, such as density, to estimate liquid thermal conductivity (Sastri and Rao, 1999). In summary, apart from a few exceptions, many estimation models neglect the pressure dependency and represent the liquid thermal conductivity as a function of temperature only.

In this paper, some of the most widespread and well-known models available in scientific literature are analyzed to estimate the thermal conductivity of low GWP refrigerants at the liquid state. Moreover, modified versions of different correlations to describe the liquid thermal conductivity of these alternative refrigerants and their dependence on both temperature and pressure are presented.

2. Data analysis

A literature survey was performed to define a reliable and updated experimental dataset of the liquid thermal conductivity for halogenated alkene refrigerants. Experimental λ_L data for 6 refrigerants were finally collected, confirming the limited number of potential low GWP working fluids for which the transport properties were measured (Bobbo et al., 2018).

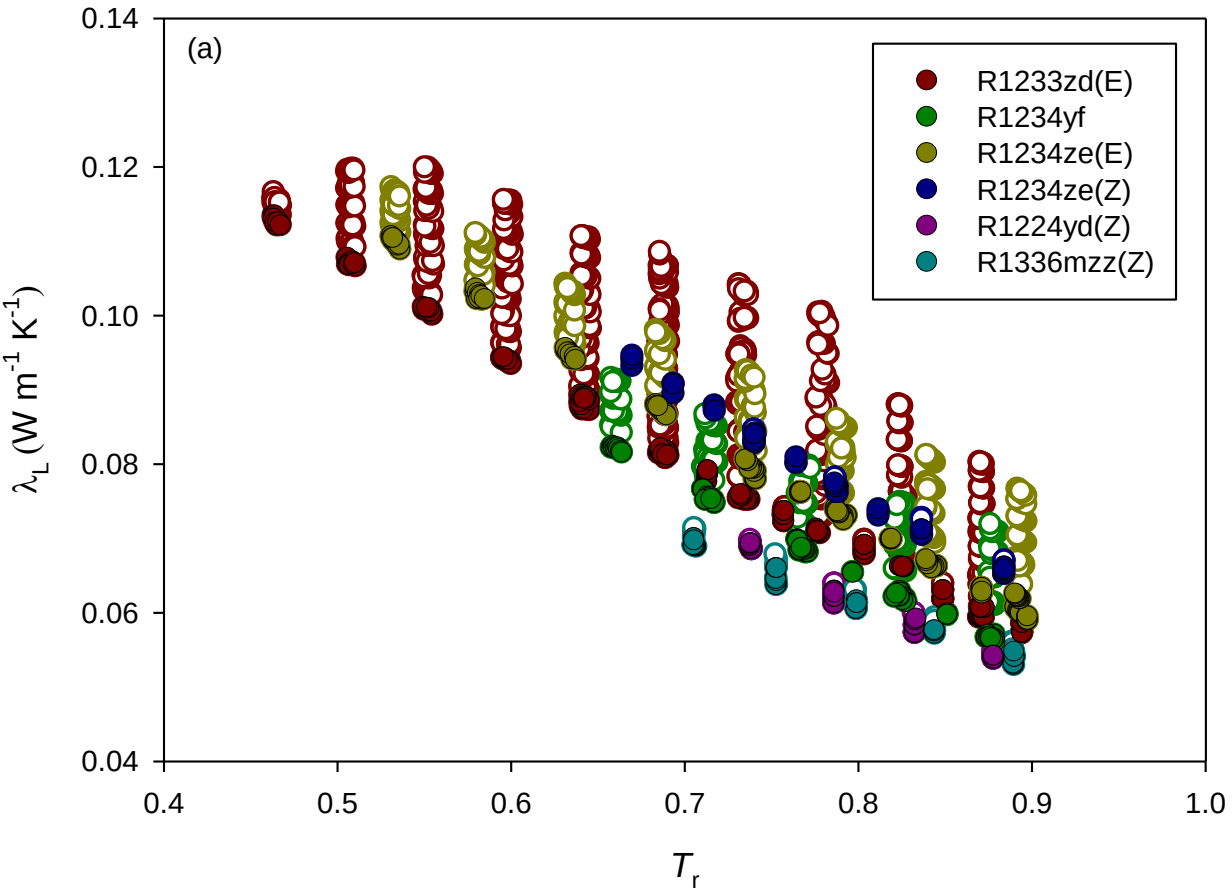
Then, we performed a critical fluid by fluid analysis and selection of the collected experimental values. The experimental data showing deviations more than three sigma from the mean values and data clearly beyond the common trend were rejected. In this regard, the values of thermal conductivity for R1234ze(E) presented by Grebenkov et al. (2009) and Miyara et al. (2010) were neglected, since they show a different behavior with reduced temperature ($T_r = T\ T_c^{-1}$, where T_c stands for critical temperature) and reduced pressure ($P_r = P\ P_c^{-1}$, where P_c stands for critical pressure) respect to the other sources. Moreover, considering the abovementioned limitations of the correlations for λ_L near the critical point, the experimental values measured up to T_r equal to 0.9 were selected.

Finally, from this selecting process, a dataset containing a total of 2073 experimental values for 6 alternative refrigerants was defined. The collected experimental data come from reliable apparatuses based on the transient hot-wire method, and experimental sources declare uncertainties

lower than 3 % in most cases. Table 1 provides the temperature, pressure and thermal conductivity ranges for the studied fluids, together with the number of points and the references of the data sources. The trends of the selected λ_L data as function of T_r and P_r are shown in Figure 1.

Table 1. Summary of the experimental liquid thermal conductivity data for the halogenated alkene refrigerants studied in this work.

Refrigerant	CAS number	Chemical formula	Number of data	T range, K	P range, MPa	λ_L range, W m^{-1} K^{-1}	Source
R1233zd(E)	102687-65-0	$C_3ClF_3H_2$	1132	203.56 - 393.22	0.18 - 66.62	0.05719 - 0.11991	Perkins et al. (2017)
							Alam et al. (2018)
R1234yf	754-12-1	$C_3F_4H_2$	267	241.92 - 324.00	0.44 - 21.64	0.05607 - 0.09158	Perkins and Huber (2011)
							Miyara et al. (2011a)
R1234ze(E)	29118-24-9	$C_3F_4H_2$	494	203.18 - 343.31	0.31 - 23.32	0.05893 - 0.11727	Perkins and Huber (2011)
							Miyara et al. (2011a)
							Miyara et al. (2011b)
R1234ze(Z)	29118-25-0	$C_3F_4H_2$	61	283.54 - 374.24	0.10 - 4.01	0.06520 - 0.09457	Ishida et al. (2015)
							Miyara (2019)
R1224yd(Z)	111512-60-8	C_3ClF_4H	53	316.25 - 376.37	1.00 - 4.07	0.05379 - 0.06979	Alam et al. (2019)
R1336mzz(Z)	692-49-9	$C_4F_6H_2$	66	313.43 - 395.70	0.54 - 4.08	0.05294 - 0.07136	Alam et al. (2017)


(a)
R1233zd(E)
R1234yf
R1234ze(E)
R1234ze(Z)
R1224yd(Z)
R1336mzz(Z)
0.14
0.12
0.10
0.08
0.06
0.04
0.4
0.5
0.6
0.7
0.8
0.9
1.0
λ_L (W m^{-1} K^{-1})
T_r

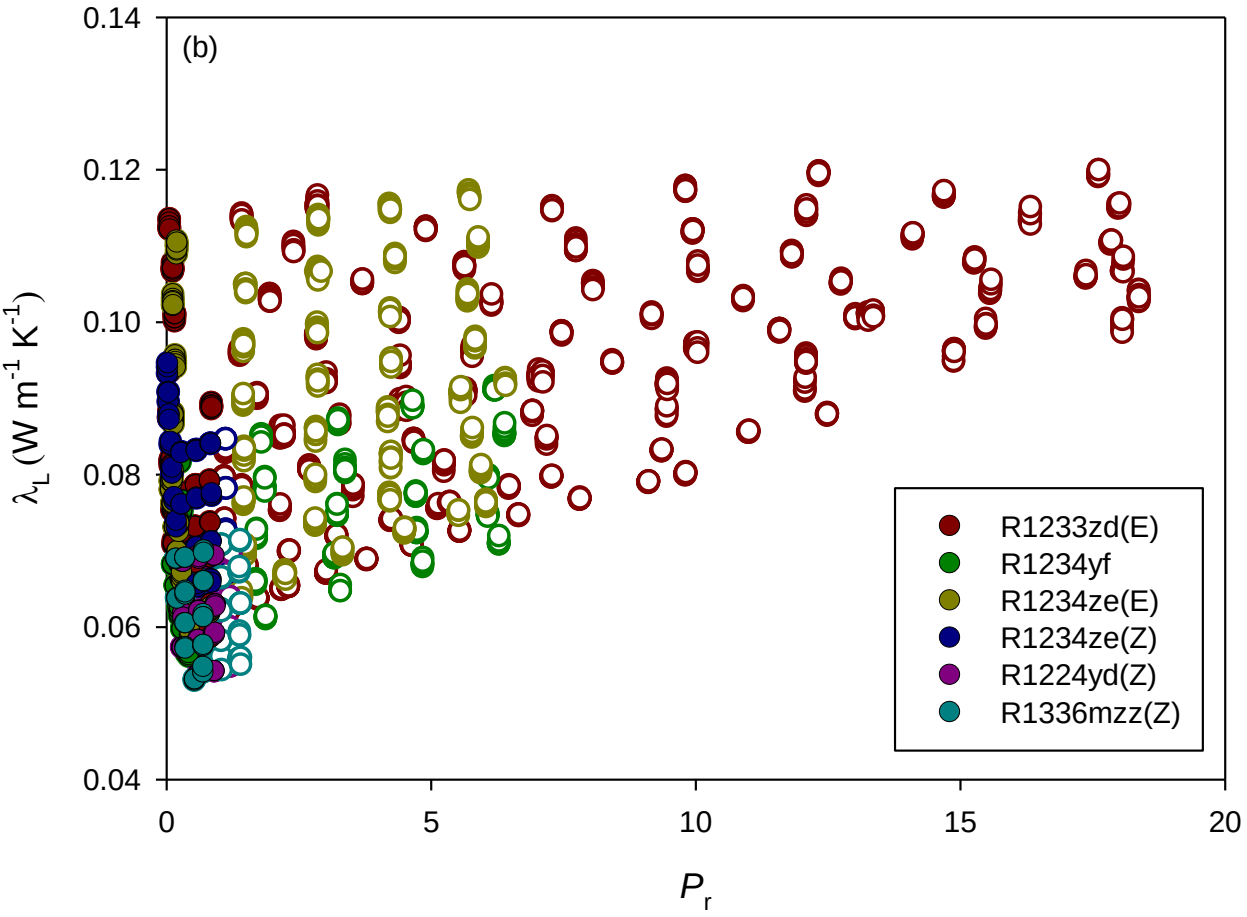


Figure 1. Behaviors of the experimental liquid thermal conductivity, λ_L, data for the halogenated alkene refrigerants as function of reduced temperature, T_r, (a) and reduced pressure, P_r, (b). The filled symbols represent the points measured at $P_r < 1$ and the open symbols represent the points measured at $P_r > 1$.

From Table 1 and Figure 1, it is possible to note that the selected experimental data were measured over the following wide liquid thermal conductivity, temperature, and pressure ranges: (0.04805 - 0.11727) W m^{-1} K^{-1}, (203.18 - 434.99) K, and (0.10 – 66.62) MPa. This aspect is even more evident in Figure 2, where the experimental T_r and P_r of the measured data are shown together with the vapor-liquid saturation boundaries of the studied refrigerants. The values of the vapor-liquid saturation boundary were calculated from the Equations of State (EoSs) of the studied fluids used in

REFPROP 10.0 (Lemmon et al., 2018), as reported in Table 2.

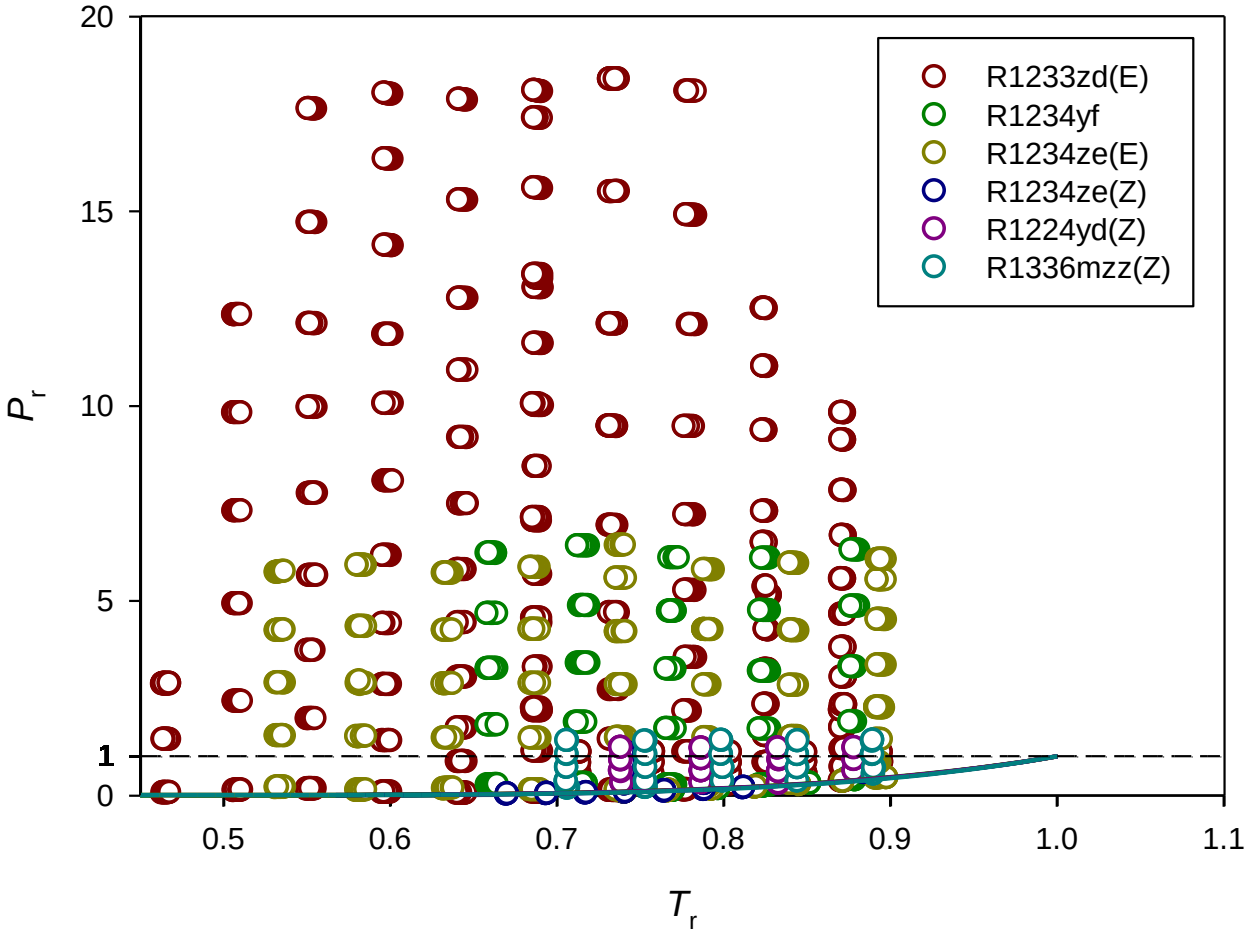


Figure 2. Reduced temperature range and reduced pressure range of the collected experimental points for the studied liquid refrigerants. The solid lines are the vapor-liquid saturation boundaries calculated from REFPROP 10.0.

Figure 1b also shows the expected thermal conductivity increment with reduced pressure. However, since its effect on λ_L is limited and thus negligible up to moderate pressures, the collected data up to $P_r = 1$ are basically constant at a fixed reduced temperature, as proved by the filled symbols in Figure 1. Table 2 reports the physical properties of the fluids analyzed in this work: molecular mass, M, normal boiling point temperature, T_b, critical temperature, T_c, critical pressure, P_c, acentric factor,

ω, and dipole moment, μ. With few exceptions pointed out in Table 2, these properties were collected from REFPROP 10.0 (Lemmon et al., 2018) and used to develop the EoSs of the studied refrigerants. Instead, the dipole moments of R1234yf and R1234ze(E), the critical temperature of R1224yd(Z), and the critical pressures of R1224yd(Z) and R1336mzz(Z) were taken from recent and reliable sources (Sampson et al., 2019, Sakoda and Higashi, 2019 and Alam et al., 2017).

Table 2. Physical properties for the selected refrigerants studied in this work.

Refrigerant	M kg kmol^{-1}	T_b K	T_c K	P_c MPa	ω	μ D	Source
R1233zd(E)	130.496	291.41	439.60	3.62	0.3025	1.12	Mondéjar et al. (2015)
R1234yf	114.042	243.67	367.85	3.38	0.2760	2.24[a]	Richter et al. (2011)
R1234ze(E)	114.042	254.18	382.51	3.63	0.3130	1.13[a]	Thol and Lemmon (2016)
R1234ze(Z)	114.042	282.88	423.27	3.53	0.3274	2.90	Akasaka and Lemmon (2019)
R1224yd(Z)	148.487	287.77	428.69[b]	3.33[b]	0.3220	1.47	Akasaka et al. (2017)
R1336mzz(Z)	164.056	306.55	444.50	2.90[c]	0.3867	2.92	Lemmon et al. (2018)

[a] Value given in Sampson et al. (2019)
[b] Value given in Sakoda and Higashi (2019)
[c] Value given in Alam et al. (2017)

3. Estimation of liquid thermal conductivity at moderate pressures

In this section, different estimation methods for low GWP refrigerants that represent the liquid thermal conductivity dependence on temperature, but not considering its dependence on pressure, are analyzed. Also, the results provided by some of these models, re-fitted for the selected refrigerants, are shown and discussed. This analysis includes only the λ_L values measured up to $P_r < 1$ (a total of 499 data points), which showed a negligible pressure dependence. Instead, the results obtained for the complete dataset, analyzed with reduced-pressure-dependent models, will be discussed in Section 4.

3.1 Literature correlations

Among the several empirical and semi-empirical estimation methods available for λ_L, only the models that complied with at least one of the following criteria were selected and analyzed in the present paper:

- the model should be specifically developed for refrigerants (or at least for a large group of families of fluids);
- only a limited number of easily available and well-known physical properties should be contained in the model;
- the coefficients of the model should be as much general as possible, i.e. they should not be dedicated to single compounds, but to different families of fluids.

On the basis of the aforementioned criteria, 4 estimation methods showing a reasonable compromise between simplicity and generality have been selected. A brief description of these models is provided below.

The Sato-Riedel equation (Poling et al., 2001) is a rather simple model that can be used for all organic fluids, including refrigerants, over a wide temperature range. This equation has the following expression:

$$\lambda_L = \frac{1.1053}{M^{1/2}} \frac{3+20\left(1-T_r\right)^{2/3}}{3+20\left(1-T_{br}\right)^{2/3}} \qquad (1)$$

where M is the molecular mass in kg kmol^{-1}, $T_r = T\,T_c^{-1}$ is the reduced temperature, T is the temperature in K, T_c is the critical temperature in K, $T_{br} = T_b\,T_c^{-1}$ is the reduced normal boiling point temperature, and T_b is the normal boiling point temperature in K.

A very well-known and widespread equation for several families of organic liquids is the Latini equation. Starting from the 70s, different expressions of this model were proposed by the author and his co-workers for several families of organic compounds (Latini et al., 2017, 2016; Latini and Pacetti, 1977). Among them, an expression presenting parameters specific for three families of refrigerants (methane, ethane, and propane series) was developed (Latini and Sotte, 2012, 2011). This equation has the following form:

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

where

$$A = A' \frac{T_b^{\alpha}}{M^{\beta} T_c^{\gamma}}, \qquad (3)$$

Φ = 1.618033… is the golden ratio, whose value is linked with the Fibonacci's sequence, and a, A', α, β, and γ are coefficients regressed for each series of refrigerants. For the refrigerants under study, the authors showed that the equation provided more accurate results than those of other models available in literature. However, the main drawback of Equation (2) is that the model requires 5 coefficients specifically given for each refrigerant series. In addition, being a linear equation with temperature, it is less accurate on the extremities of the analyzed ranges.

To overcome the limitations of some literature models and to estimate accurate λ_L near the critical point, Gharagheizi et al. (2013) developed a specific equation using a Gene Expression Programming (GEP) mathematical strategy (Ferreira, 2001), defined as:

$$\lambda_{\mathrm{L}} = 1\cdot 10^{-4}\left[10\omega + 2P_{\mathrm{c}} - 2T + 4 + 1.908\left(T_{\mathrm{b}} + \frac{1.009B^2}{M^2}\right) + \frac{3.9287M^4}{B^4} + \frac{A}{B^8}\right] \quad (4)$$

where

$$A = 3.8588M^8\left(1.0045B + 6.5152M - 8.9756\right), \quad (5)$$

$$B = 16.0407M + 2T_{\mathrm{b}} - 27.9074, \quad (6)$$

ω is the acentric factor, and P_{c} is the critical pressure in bar. This equation was developed to cover a wide number of chemical families, including amines, silanes, nitriles, inorganic compounds, elements, and aldehydes. However, the main issue of this empirical model is the high number of regressed coefficients.

Among the different estimation models of λ_{L} for organic compounds, such as CSP (Di Nicola et al., 2014b) and artificial neural networks (Di Nicola et al., 2016; Pierantozzi and Petrucci, 2018), a simple empirical correlation for liquid refrigerants with four parameters was presented by Di Nicola et al. (2014a). This equation has the following form:

$$\frac{\lambda_{\mathrm{L}}}{\lambda_0} = aT_{\mathrm{r}} + bP_{\mathrm{c}} + c\omega + \left(\frac{1}{M}\right)^d \quad (7)$$

where the original coefficients λ_0, a, b, c, and d provided by Di Nicola et al. (2014a) are reported in Table 3 (namely Equation (7)a). In addition, the authors analyzed the influence of the dipole moment on λ_{L} and provided an updated version of Equation (7); in this case, the coefficients are given for the complete dataset and for four refrigerants subgroups, defined according to the different chemical halogens forming the compounds. The new equation is defined as:

$$\frac{\lambda_{\mathrm{L}}}{\lambda_0} = aT_{\mathrm{r}} + bP_{\mathrm{c}} + c\omega + \left(\frac{1}{M}\right)^d + e\mu \quad (8)$$

where the original values of λ_0, a, b, c, d, and e for the complete dataset are reported in Table 3. It is worthwhile noting that the coefficients of Equation (7)a and Equation (8) were regressed on the data

of refrigerants of different generations. Therefore, despite their chemical and engineering interest, they are not specifically oriented to alternative refrigerants.

Table 3. Original coefficients of Equation (7)a and Equation (8) from Di Nicola et al. (2014a) and coefficients proposed in this work for Equation (7)b and Equation (12).

Eq.	λ_0 W m^{-1} K^{-1}	a	b bar^{-1}	c	d	e D	f_0	f	g
(7)a	0.5147	-0.2537	0.0017	0.1501	0.2999	-	-	-	-
(8)	0.6542	-0.2034	0.0013	0.1714	0.3539	-0.0070	-	-	-
(7)b	0.43693	-0.28725	0.00372	0.26967	0.36436	-	-	-	-
(12)	0.43693	-0.28725	0.00372	0.26967	0.36436	-	-0.00135	0.05484	0.88049

3.2 Modified versions of correlations

To improve the estimation capability of λ_L for the studied low GWP refrigerants, a modified version of Equation (7) proposed by Di Nicola et al. (2014a) has been developed. In particular, the optimized coefficients of this correlation were determined through a Random Search method (Andradóttir, 2006), available in Wolfram Mathematica, by minimizing the Average Absolute Relative Deviation of the liquid thermal conductivity, AARD(λ_L), between the selected experimental dataset and the calculated values. The AARD(λ_L) is expressed as:

$$AARD(\lambda_L) = \frac{100}{N}\sum_{i=1}^{N}\frac{\left|\lambda_{L,exp,i} - \lambda_{L,calc,i}\right|}{\lambda_{L,exp,i}} \qquad (9)$$

where $\lambda_{L,exp}$ is the experimental liquid thermal conductivity, $\lambda_{L,calc}$ is the calculated thermal conductivity, and N is the number of experimental points. The physical properties given in Table 2 were used for the regression of the parameters reported in Table 3 (indicated as Equation (7)b).

Since the coefficients of Equation (2) developed by Latini and Sotte (2012) were calculated only for three refrigerants series (methane, ethane, and propane series), the λ_L estimations provided by this equation with the available coefficients are not reliable for the low GWP refrigerants under study.

For this reason, the coefficients of Equations (2) and (3) were regressed minimizing the AARD(λ_L) with the Random Search method. By using the physical properties of Table 2, the following expression was obtained for the studied six alternative refrigerants:

$$\lambda_L = 0.066 \frac{T_b^{1.877}}{M^{0.811} T_c^{0.989}} \left[\sqrt{5} \frac{\left(\Phi - T_r\right)^2}{\Phi - T_r} \right]^{0.6} \qquad (10)$$

3.3 Results and discussion

The results provided by Equation (7)b and Equation (10) are shown in Table 4, and Figure 3 and 4. Additionally, Table 4 reports the AARD(λ_L) and the Maximum Absolute Relative Deviations for λ_L, MARD(λ_L), for the literature estimation methods analyzed in this study. From Table 4, it is evident that the correlations previously presented by our group, namely Equation (7)a and Equation (8), perform quite well even when the originally proposed coefficients are used. This is true for almost all the fluids, excluding R1234yf for which deviations are much higher. The results provided by Equation (7)b and Equation (10) overcome this issue, showing deviations of, respectively, 0.88 % and 2.47 % for this important HFO. In general, Equations (7)b and (10) present very low overall deviations, namely 1.78 % and 2.39 %.

Figure 5 shows the deviations between the calculated and experimental λ_L data for the selected refrigerants. From Figure 5 and Table 4, it is evident that the highest deviations are provided by Equation (1) and Equation (4). This is probably due to the fact that they are general correlations developed for a large group of families of fluids, and not specifically oriented to refrigerants.

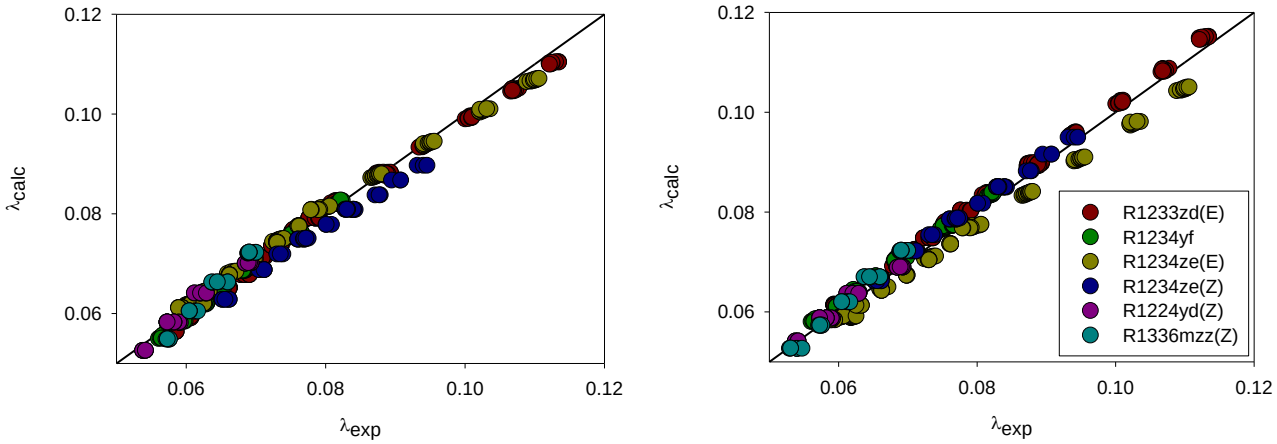


Figure 3. Liquid thermal conductivity calculated with Equation (7)b (left) and Equation (10) (right) versus experimental liquid thermal conductivity data up to P_r =1.

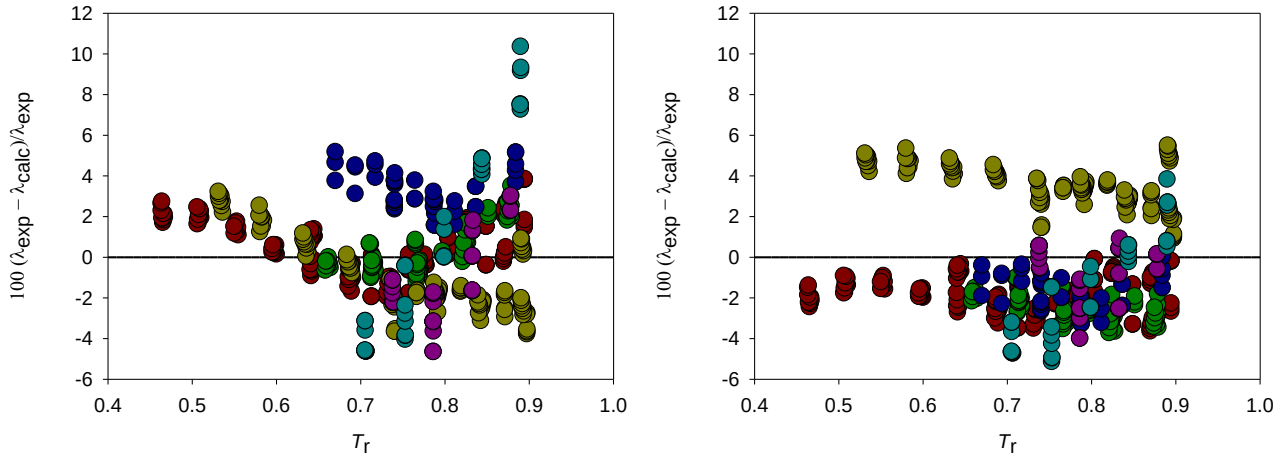


Figure 4. Deviations between liquid thermal conductivity measurements up to P_r = 1 and values calculated with Equation (7)b (left) and Equation (10) (right) versus the reduced temperature. Notation as in Figure 3.

Table 4. Average Absolute Relative Deviations, AARD(λ_L) %, and Maximum Absolute Relative Deviations, MARD(λ_L) %, between liquid thermal conductivities, λ_L, values provided by Equation (1), Equation (4), Equation (7)a, Equation (8), Equation (7)b, and Equation (10) and the experimental data measured in the reduced pressure range up to 1.

Fluids	N. points	Eq. (1)	Eq. (4)	Eq. (7)a	Eq. (8)	Eq. (7)b	Eq. (10)
		AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %
		MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %
R1233zd(E)	161	10.09	5.57	2.33	2.38	1.20	1.85
		15.01	12.26	4.89	4.92	3.85	3.64
R1234yf	82	22.94	9.68	8.53	1.58	0.88	2.47
		27.14	19.14	10.10	3.66	3.52	3.72
R1234ze(E)	131	10.22	6.69	4.46	5.72	1.83	3.53
		16.83	14.31	7.29	8.63	3.77	5.49
R1234ze(Z)	49	7.83	3.18	0.87	8.17	3.29	1.70
		11.13	7.17	2.10	12.09	5.18	3.27
R1224yd(Z)	40	14.47	11.88	4.30	2.56	1.98	1.33
		20.09	15.19	8.22	5.46	4.65	4.00
R1336mzz(Z)	36	14.22	11.25	4.94	8.01	3.94	2.56
		21.82	13.32	8.08	19.96	10.35	5.14
Overall	499	12.66	7.22	4.11	4.12	1.78	2.39
		-	-	-	-	-	-

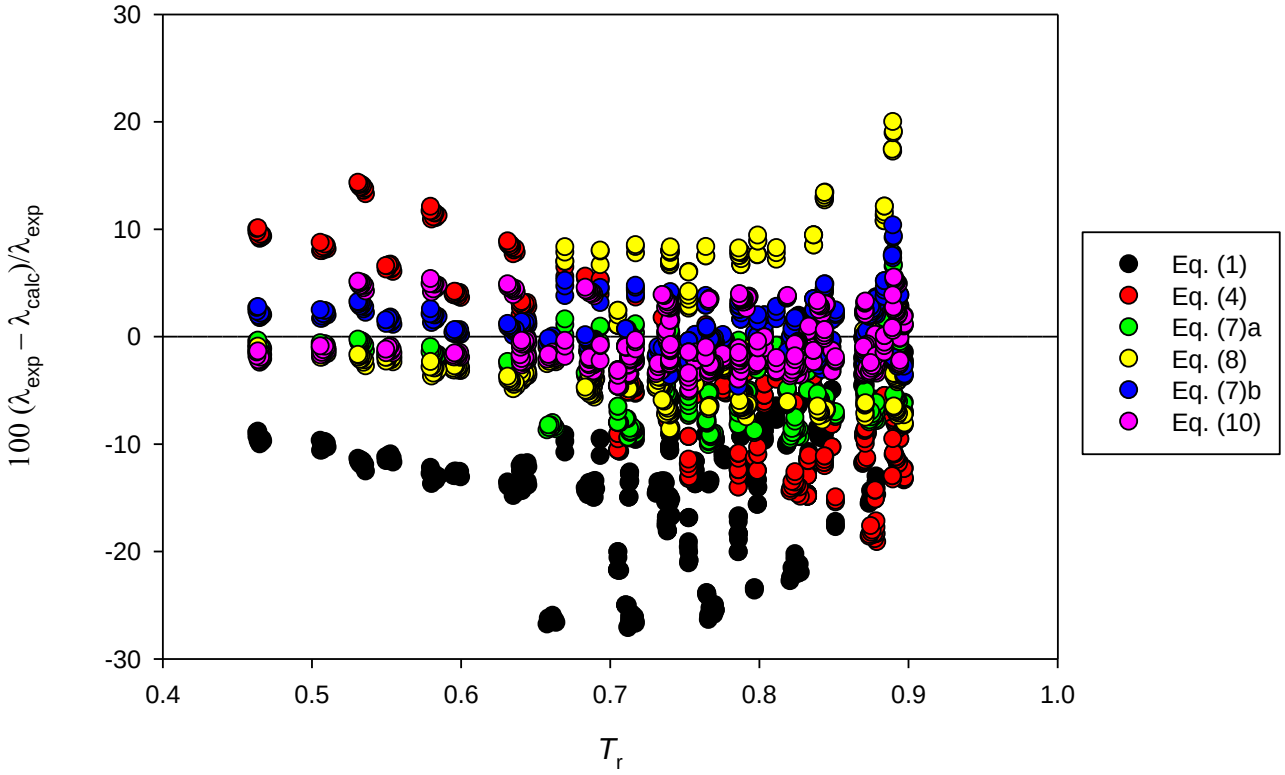


Figure 5. Deviations between liquid thermal conductivity measurements up to P_r = 1 and values calculated from all the studied models versus the reduced temperature.

4. Pressure-dependent models

In this section, correlations for λ_L that take into account the pressure dependence are analyzed for the experimental dataset of the studied alternative refrigerants. Although a slight λ_L dependence on pressure is evident from the data shown in Figure 1, Equation (7)a gave an AARD(λ_L) equal to 7.15 % for all the 2073 experimental data and, in particular, it gave MARD(λ_L) = 27.61 % for R1233zd(E). Compared to the lower deviations reported in Table 4, these results show that Equation (7)a is not adequate to estimate λ_L at all temperature and pressure conditions. In the same fashion, Equation (7)b and Equation (10) gave AARD(λ_L) equal to 11.02 % and 10.30 %, respectively, for the data measured at $P_r \geq 1$ (1574 data points). Therefore, pressure-dependent versions of the models described above were developed and analyzed. Their results are reported below and compared with the values provided

by a pressure-depended correlation published recently (Amooey, 2017) and REFPROP 10. (Lemmon et al., 2018). REFPROP 10.0 provides the thermal conductivity values using either fluid-specific correlations or a modification of the extended CSP method. In particular, these models describe the thermal conductivity dependence on temperature and pressure at different thermodynamic conditions.

A limited number of correlations describing the pressure dependence of λ_L for high values have been proposed in literature (Poling et al., 2001). Missenard (1970) developed the following expression to estimate pressure-dependent λ_L up to very high pressures:

$$\frac{\lambda_L(P_r)}{\lambda_L(\text{low } P)} = 1 + QP_r^{0.7} \qquad (11)$$

where $\lambda_L(P_r)$ and $\lambda_L(\text{low } P)$ are the liquid thermal conductivities at high and low (near saturation) pressures, respectively, at a fixed temperature, while Q is a coefficient defined for different values of T_r and P_r.

Based on the dependence of P_r with T_r described in Equation (11), a modified version of Equation (7) that takes into account the effect of pressure as a function of T_r has been developed and analyzed. The correlation that was found to ensure the best results has the following form:

$$\frac{\lambda_L(P_r)}{\lambda_0} = \left[aT_r + bP_c + c\omega + \left(\frac{1}{M}\right)^d \right] \left[1 + \left(f_0 + fT_r^2 \right) P_r^g \right] \qquad (12)$$

It is important noting that Equation (12) has to be considered an extension of Equation (7)b, which was found to be the best option amongst the pressure-independent models. For this reason, the original coefficients λ_0, a, b, c and d found for Equation (7)b and reported in Table 3 were kept constant while only the coefficients f_0, f, and g were determined through the Random Search method (Andradóttir, 2006) by minimizing the AARD(λ_L) for the complete dataset. The values of f_0, f, and g are also reported in Table 3.

To include the effect of pressure in their correlation, Latini and his coworkers (Latini et al., 1989; Latini and Baroncini, 1983) modified the expression of the parameter A as:

$$A = A_0 + A_1 P_r \qquad (13)$$

where A_0 corresponds to the low pressure parameter, such as the one defined by Equation (3) for refrigerants, and A_1 is a parameter that was generalized for hydrocarbons as:

$$A_1 = \frac{\alpha_1}{M^{\beta_1}} \qquad (14)$$

where coefficients α_1 and β_1 were provided for saturated hydrocarbons and aromatics.

In this study, an updated version of Equation (10) that includes the λ_L dependence on pressure, and that is also a function of T_r, has been assessed. The new coefficients were regressed using the Random Search method (Andradóttir, 2006) by minimizing the AARD(λ_L) for the complete dataset. The following expression was obtained:

$$\lambda_L(P_r) = \left[0.066 \frac{T_b^{1.877}}{M^{0.811} T_c^{0.989}} + \frac{1}{M^{(1.857-0.755T_r)}} P_r\right]\left[\sqrt{5}\frac{(\Phi - T_r)^2}{\Phi - T_r}\right]^{0.6} \qquad (15)$$

More recently, Amooey (2017) presented a correlation to estimate λ_L of refrigerants over wide ranges of temperature and pressures. The new model was developed considering the data for 20 liquid refrigerants, among which R1234yf and R1234ze(E), that were divided into three subgroups based on the different chemical halogens forming the compounds, namely Cl- derivatives, F- derivatives, and F + Cl + Br- derivatives. The proposed equation is expressed as:

$$\frac{\lambda_L(P_r)}{\lambda_0} = B_1 T_r + B_2 P_r + B_3 \omega + \left(\frac{B_4}{M}\right)^{B_5} \qquad (16)$$

where the coefficients B_1, B_2, B_3, B_4, and B_5, were regressed for the three subgroups individually.

The deviations between all the collected experimental λ_L data and the values provided by Equation (12) and Equation (15) are shown in Figure 6. Equation (12) and Equation (15) are the pressure-dependent correlations proposed in this paper. From Figure 6, it is evident that both models perform well, being almost all the deviations within ± 6 %. Figure 7 also compares the proposed models together with the results calculated according to Equation (16) for R1234yf. Based on Figure 7, it is possible to note that the proposed correlations accurately describe the λ_L dependence on pressure

for R1234yf, especially at lower reduced temperatures. Instead, Equation (16) provides less accurate estimations for the same refrigerant.

The AARD(λ_L) and MARD(λ_L) for Equation (12), Equation (15), Equation (16), and REFPROP 10.0 are reported in Table 5. The table shows the overall good results obtained by the proposed models (deviations of 1.45 % and 1.99 % for Equation (12) and Equation (15), respectively), even when a limited number of data is available. Very low deviations (0.54 %) are obtained when REFPROP 10.0 is used to calculate the liquid thermal conductivity. REFPROP 10.0 can be considered as a reference, since it calculates thermal conductivity using fluid-specific correlations for all the refrigerants under analysis except R1224yd(Z), for which a completely predictive model was used (Huber, 2018). In fact, for this fluid REFPROP 10.0 provided less accurate λ_L estimations (AARD(λ_L) = 6.36 %). Finally, it was checked that the proposed pressure-dependent correlations ensure results similar to that of Equation (7)b and Equation (10) for the data measured at $P_r < 1$ (overall AARD(λ_L) of 1.99 % and 2.85 % for Equation (12) and Equation (15), respectively).

Considering the similarity between the pressure-volume-temperature and temperature–thermal conductivity–pressure diagrams for fluids, models based on EoSs were developed to estimate λ of pure fluids and mixtures in different phase regions. In particular, Khosharay et al. (2018) presented a thermal conductivity model based on Heyen's EoS (Heyen, 1980) for 31 conventional refrigerants (none of them was analyzed in this paper) and their mixtures, that can be used for liquid, vapor and supercritical regions. Although this model only needs thermal conductivity, pressure, and temperature information as input parameters, and it is applicable for both low and high pressures, it uses coefficients regressed for each refrigerant. For all these reasons, a comparison between this model and the proposed correlations could not be performed.

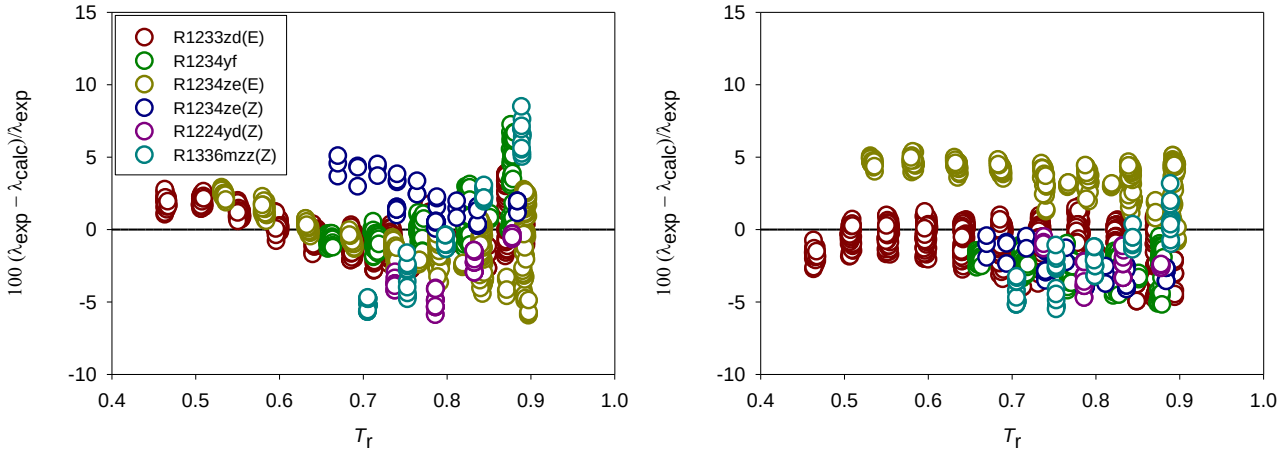
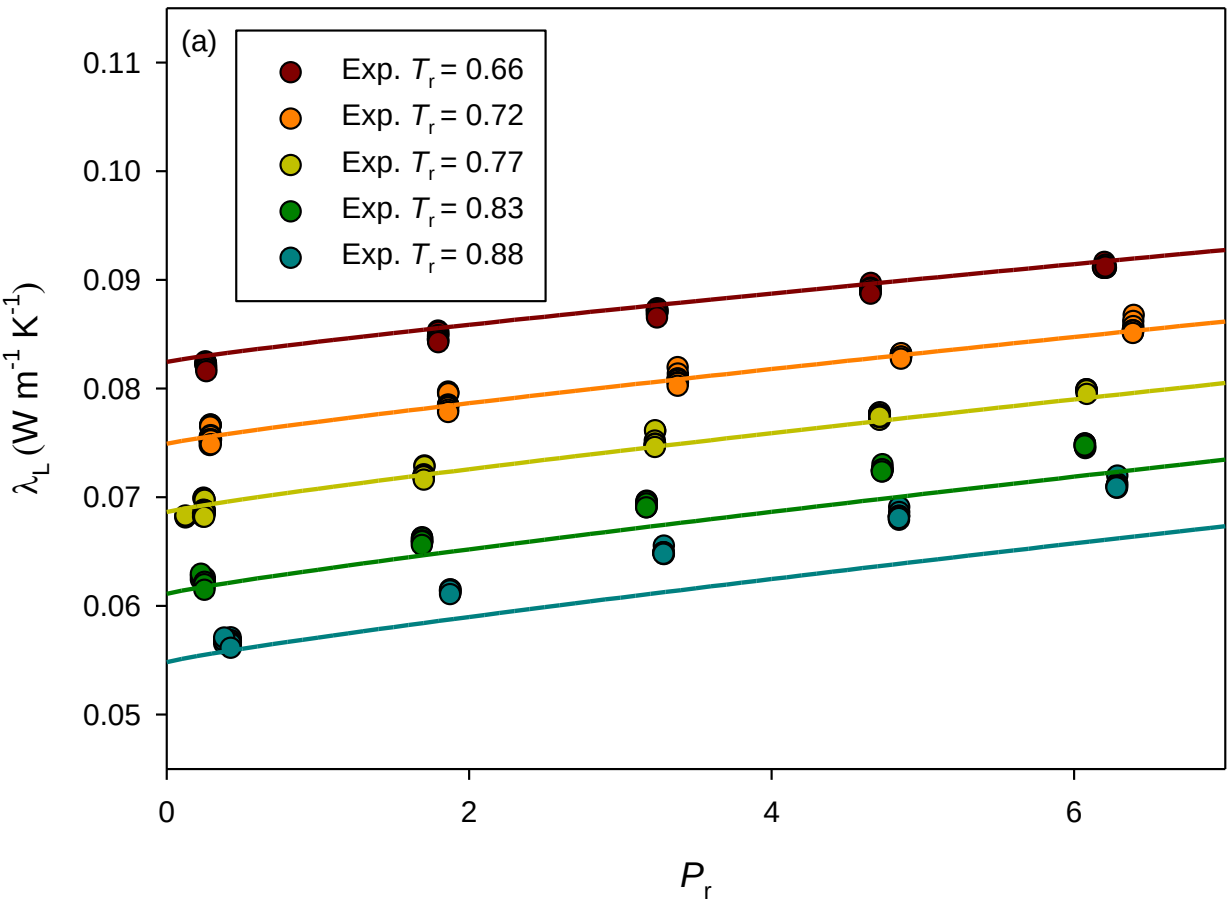
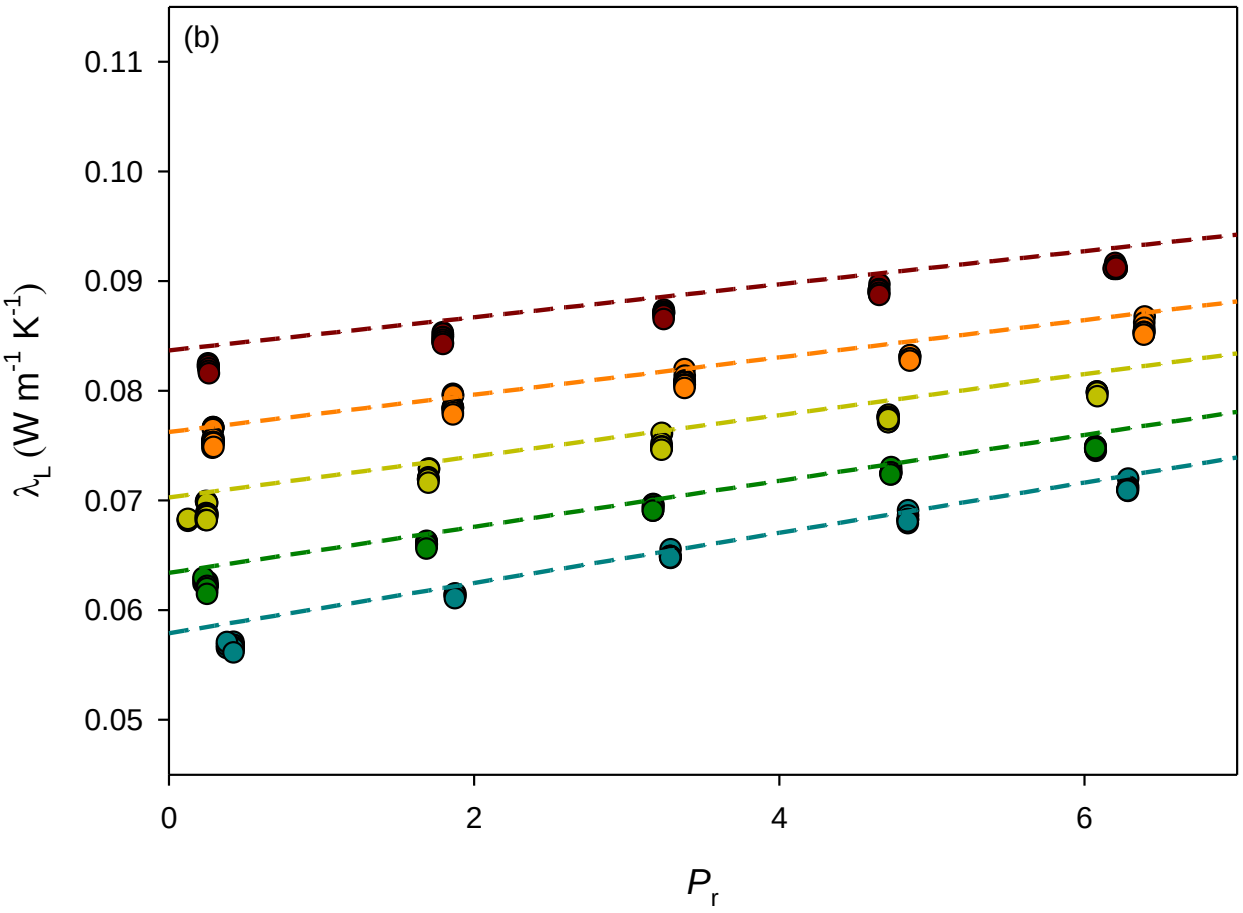


Figure 6. Deviations between the collected experimental liquid thermal conductivity data and values calculated from Equation (12) (left) and Equation (15) (right) versus reduced temperature.


(a)
Exp. T_r = 0.66
Exp. T_r = 0.72
Exp. T_r = 0.77
Exp. T_r = 0.83
Exp. T_r = 0.88
λ_L (W m^{-1} K^{-1})
0.11
0.10
0.09
0.08
0.07
0.06
0.05
0
2
4
6
P_r


(b)
0.11
0.10
0.09
0.08
0.07
0.06
0.05
λL (W m-1 K-1)
0
2
4
6
Pr

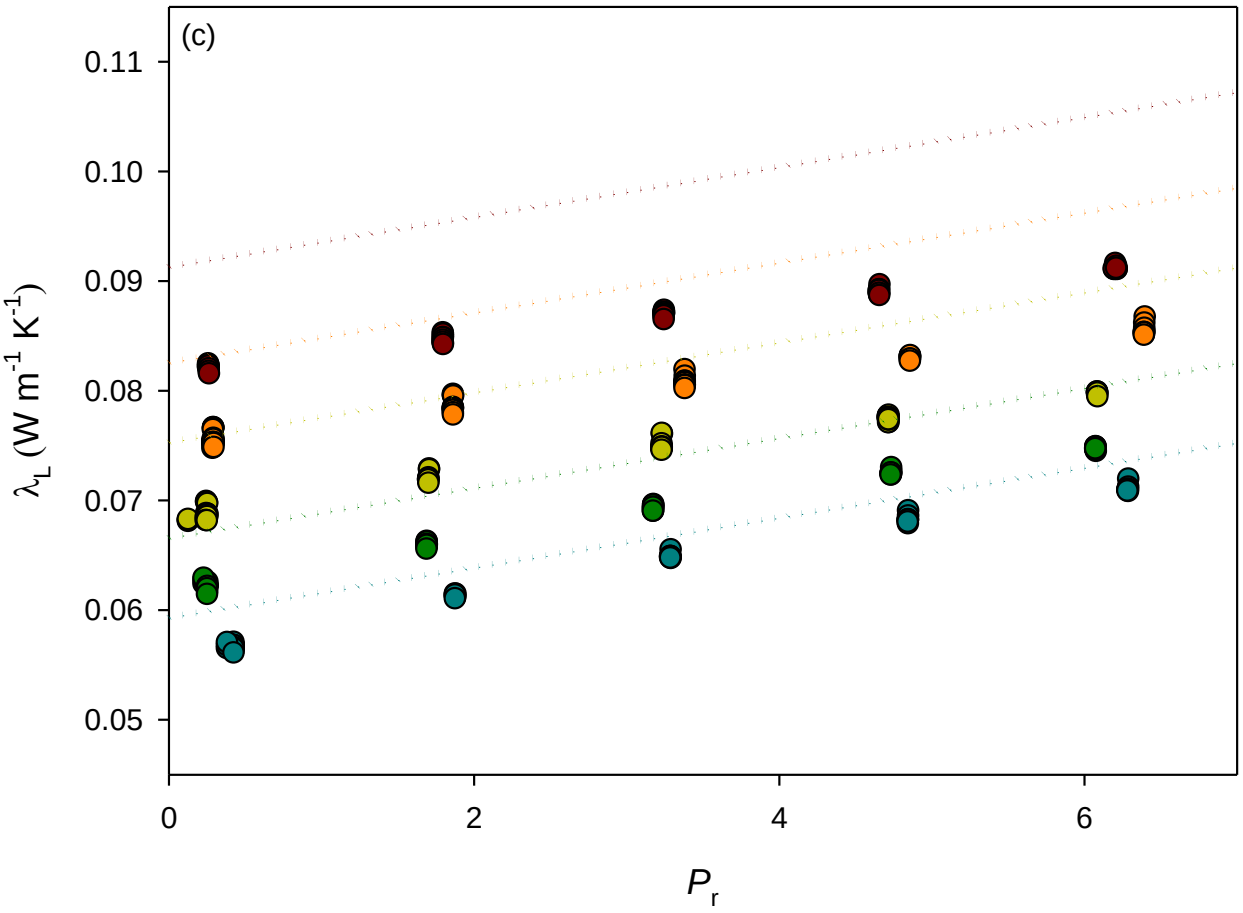


Figure 7. Experimental liquid thermal conductivity data for R1234yf and values calculated with Equation (12) (solid line) (a), Equation (15) (short dash line) (b), and Equation (16) (dotted line) (c) versus reduced pressure for the collected reduced temperatures.

Table 5. Average Absolute Relative Deviations, AARD(λ_L) %, and Maximum Absolute Relative Deviations, MARD(λ_L) %, between liquid thermal conductivities, λ_L, values provided by Equation (12), Equation (15), Equation (16), and REFPROP 10.0 and the collected experimental data.

Fluids	N. points	Eq. (12)	Eq. (15)	Eq. (16)	REFPROP 10.0
		AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %	AARD(λ_L) %
		MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %	MARD(λ_L) %
R1233zd(E)	1132	1.15	0.98	7.26	0.34
		3.93	4.96	17.93	1.58
R1234yf	267	1.45	2.45	10.27	0.30
		7.24	5.19	16.05	1.56
R1234ze(E)	494	1.63	3.86	9.43	0.34
		5.94	5.38	18.20	2.04
R1234ze(Z)	61	1.77	2.81	18.42	1.78
		5.08	4.13	24.60	5.70
R1224yd(Z)	53	3.04	2.39	3.37	6.36
		5.90	4.72	7.42	8.86
R1336mzz(Z)	66	3.64	2.50	32.88	0.70
		8.48	5.50	48.93	2.17
Overall	2073	1.45	1.99	9.21	0.54
		-	-	-	-

5. Conclusions

In this paper, the Di Nicola et al. (2014a) correlation and the Latini and Sotte (2012) correlation were re-fitted to describe the liquid thermal conductivity dependence on temperature for 6 low global warming potential refrigerants at reduced temperatures up to 0.9. The re-fitted Di Nicola et al. (2014a) correlation (AARD(λ_L) = 1.78 %) and Latini and Sotte (2012) correlation (AARD(λ_L) = 2.39 %) provided results more accurate than those of the original versions. Also, they proved to be better than other literature estimation models for the selected data at reduced pressure lower than 1.

Moreover, the two correlations were modified to describe the temperature and pressure dependence of liquid thermal conductivity in the wide temperature and pressure ranges of the collected data. The pressure-dependent versions of the correlations ensured accurate estimations for the refrigerants under study: we found an overall AARD(λ_L) = 1.45 % for the refitted pressure-dependent Di Nicola et al. (2014a) correlation, and an overall AARD(λ_L) = 1.99 % for the pressure-dependent Latini and Sotte (2012) correlation.

Based on the results obtained for the refrigerants under study, the refitted pressure-dependent Di Nicola et al. (2014a) correlation is recommended for all the temperature and pressure conditions. At near-saturation pressures, however, the pressure-dependent term can be neglected. The proposed model can be very useful to estimate the liquid thermal conductivity of the fluids covered in this work, in particular for regions where experimental data are not available.

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