

A Prediction Method for Thermal Conductivity of Alternative Refrigerants in the Liquid Phase¹

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A prediction method for the thermal conductivity of halocarbon refrigerants in the saturated liquid state in the reduced temperature range 0.3 to well above 0.9 is presented in this paper. The aim of the method is to present a very simple calculation of the transport property useful for engineering purposes. The method determines thermal conductivity as a sole function of the reduced temperature and requires the knowledge of a parameter dependent upon easily available physical constants characteristic of each compound. The method is validated against experimental data available in the literature, giving average absolute deviations which are usually less than 5%, with maximum absolute deviations generally less than 10%. An extension of the method to estimate thermal conductivity of binary mixtures is also presented, along with a comparison with the few experimental data available in the literature.

KEY WORDS: halocarbon refrigerants; mixtures; prediction method; pure fluids; saturated liquid state; thermal conductivity.

1. INTRODUCTION

A prediction method to estimate the thermal conductivity of halocarbon refrigerants, also known as CFCs (chlorofluorocarbons), HCFCs (hydrochlorofluorocarbons), and HFCs (hydrofluorocarbons), in the liquid phase along the saturation line and in the subcooled state close to the saturation pressure is presented in this paper. The new method is based on earlier investigations [1-4] concerning the liquid thermal conductivity of organic compounds in the reduced temperature range 0.3 to 0.8 and represents a remarkable improvement of the old method.

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The equation proposed in this work is semiempirical in nature and has the form $\lambda = \lambda(T_r)$ where λ is the thermal conductivity in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, T_r is the reduced temperature, T/T_c , T is the temperature in K, and T_c is the critical temperature in K.

It requires the knowledge of easily available physical constants, such as the molecular weight, the critical temperature and the critical pressure, in addition to a constant which is characteristic of the organic family and dependent on the number of carbon atoms.

According to a distinction between prediction and estimation as reported in Ref. 5, the present method could be defined as an empirical estimation method because it does not appear clearly connected to a well-grounded theoretical basis. Nevertheless, the name "prediction" is maintained for the method because its simple structure allows the user to calculate the transport property, without knowledge of any experimental datum on the property itself. The predictive power of the equation has been checked by testing the method with halocarbon refrigerants not used to obtain the correlation and its parameters.

An important feature of the method is that it is simple and easy to use and is, therefore, a handy tool for many engineering calculations involving heat and mass transfer; in this case, the required knowledge of only easily available physical parameters helps considerably.

As far as the possible errors in the thermal conductivity calculation are concerned, namely, accuracy and simplicity being the two fundamental tasks, the method looks for satisfying both: it means that average absolute deviations generally less than 5% and maximum absolute deviations generally less than 10% are considered satisfactory, provided that the method is proposed for engineering purposes and the comparison is made taking into account experimental thermal conductivity values due to different authors who often use different techniques.

The range of validity of the method in reduced temperature is 0.30 to well above 0.90; it applies to pure fluids and can be used, with an appropriate mixing rule, for binary mixtures also.

The equation is validated against available experimental data which are still growing in number. The old CFCs and their alternatives are taken into account in this paper because the investigation is concerned in general with the thermal conductivity of the organic compounds in the saturated liquid phase and, from this point of view, the proposed equation must be verified by all the compounds of the same family.

2. PREVIOUS WORK ON THE METHOD

The departure point for the method was an equation proposed earlier [1,3] for pure fluids, having the form

$$\lambda = A \frac{(1 - T_r)^{0.38}}{T_r^{1/6}} \quad (1)$$

where A is a constant. It was, at the beginning, determined from experimental data of many organic liquids along the saturation line or at pressures near the saturation. Then the curve representing the experimental points and the curve corresponding to Eq. (1) were compared: it was noted that the intersection points between the two lines generally occur at $T_r \cong 0.3$, $T_r \cong 0.8$, and near $T_r \cong 0.5488$ [which is the inflection point of the function represented by Eq. (2)]. It was therefore clear that a very accurate estimation (within $\pm 2\%$) is possible by just knowing a sole experimental datum at around $T_r \cong 0.55$: the A factor obtained from that datum allows the estimation of λ for the whole $0.3 - 0.8$ reduced temperature range with great accuracy.

The search for the prediction method continued by realizing that the factor A is temperature independent to a very good approximation is characteristic of each compound and is dependent on the molecular structure of the substance. Without abandoning simplicity, a link between the factor A and the main physical properties was then detected as $A = f(M, T_b, T_c)$, where M is the molecular weight and T_b is the normal boiling temperature (at 101.325 kPa) in K.

The appropriate form of the expression for the parameter A , different from family to family (144 different compounds grouped as paraffins, organic acids, alcohols, ketones, esters, ethers, aromatics, cycloparaffins, olefins, and chlorofluorocarbons were taken into account), was then to be determined; it was found that, for the CFC refrigerants, no significant dependence on the normal boiling temperature exists and the most suitable expression for the factor A is

$$A = A^* \frac{T_c^{1/6}}{M^{1/2}} \quad (2)$$

where A^* is a constant, whose value is 0.494 for the fully halogenated methane and ethane refrigerants, namely, R10 to R14 and R110 to R160, and 0.562 for the methane-based refrigerants having a single hydrogen, namely, R20 to R23.

The method also works well when applied to HCFCs and to HFCs, as shown in Ref. 4, with average absolute deviations ranging from 2.3 to 11.1% and maximum absolute deviations ranging from 4.0 to 14.4%.

The validation of Eqs. (1) and (2) shown in Ref. 1 was developed by taking into account the experimental thermal conductivity data available up to 1980: the compounds investigated were several, but the experimental values for each compound were not many (seven or eight experimental points at the most, usually two or three points and sometimes a datum around 20°C and at atmospheric pressure) and generally were obtained by different and not comparable techniques. It is obvious, considering the situation at that time, that, for example, the exponent 0.38 in Eq. (1) comes from an average of a great number of substances; that average allowed an excellent estimation of thermal conductivity, but surely affected the choice of the value of the second exponent, i.e., 1/6. In conclusion, it is necessary to reconsider the problem of a liquid thermal conductivity prediction method for engineering purposes: this goal is possible by using a large number of selected experimental λ data for each compound over large temperature ranges. The starting point is Eq. (1) and the first group of investigated compounds is the group of the refrigerant fluids.

3. PRESENT METHOD

The previous method can be changed and substantially improved, maintaining both accuracy and simplicity, and the reduced temperature range of validity can be extended surely up to 0.90 and sometimes further. For the sake of clarity, it must be pointed out that the critical enhancement is not covered by the present method. The new coefficients and parameters appearing in the equations address the whole halocarbon refrigerants series (of methane and ethane) rather than single groups of refrigerants.

The experimental thermal conductivity values used for an extensive analysis are from different authors; they appeared in the scientific and technical literature up to 1993 and were generally obtained with different techniques; a preliminary investigation was made about the reliability of the various measurement techniques and experimental λ data were accepted which had an estimated accuracy within $\pm 5\%$. This choice was based on the following reasons:

- Even if the techniques are different and presumably affected by different sources of error, it is not possible and substantially not correct to select one author and neglect another.
- Even when the experimental thermal conductivity data due to a particular author are surely recognized as more reliable than the ones obtained by another author, often some compounds investigated by the latter are not investigated by the former.

- In some cases the estimated (or even the claimed) accuracy of experimental λ data due to a particular author is nearly $\pm 5\%$ (or slightly greater), but the investigated temperature range is very large or, in any case, the measurements are performed at temperatures not covered by other authors.

A consequence of this is that the analysis sometimes involves two thermal conductivity experimental data (for the same compound and at the same temperature) whose difference is greater than the mutual uncertainty.

Though the disadvantages of such an analysis are very clear, it is impossible to avoid the approach outlined above: the goal is a general, simple, and accurate method for saturated liquid thermal conductivity prediction and the basis and the tool of the investigation are the existing experimental thermal conductivity values. This is also another reason for the inclusion of the old refrigerants in this study.

Following an extensive work of comparison with experimental data, it was found that an excellent form for the mathematical relation between saturated liquid thermal conductivity and reduced temperature is

$$\lambda = A \left[\frac{1}{T_r} - 1 \right]^{1/3} \quad (3)$$

In Eq. (3), an exponent appears with the simple value $1/3$ and the factor A , a characteristic of each compound, is temperature independent to a very good approximation.

Equation (3) constitutes the basis for a significant check.

- If the A value is calculated from experimental data and is inserted in Eq. (3), then the average absolute deviations between estimated and experimental thermal conductivity values are generally less than 4% in the reduced temperature range from the freezing point to $T_r = 0.90$ and above.
- For such cases, in the same reduced temperature range, the maximum absolute deviations are usually less than 8%.
- At reduced temperatures close to 1.0, Eq. (3) is useless because the function $[1/T_r - 1]^{1/3}$ reaches the zero value quickly as temperature approaches the critical point, while the thermal conductivity goes to infinity at the critical point.
- The use of Eq. (3) produces the best results around the value $T_r = 2/3$, which represents the inflection point of the function $A[1/T_r - 1]^{1/3}$.

A further step is possible by writing a linear expression of the type

$$\lambda = 4^{1/3} A (1 - \frac{3}{4} T_r) = B (1 - \frac{3}{4} T_r) \quad (4)$$

which represents the equation of the tangent of the function given by Eq. (3) at the inflection point $T_r = 2/3$. The constant $B = 4^{1/3} A$ obviously has the same characteristics as the constant A . It must be noted that the behavior of Eqs. (3) and (4) is completely independent of the particular substance.

Equation (4) presents the following advantages.

- It represents a straight line and it is well-known from experimental evidence that, before the critical enhancement commences, the liquid thermal conductivity is approximately a linear function of temperature.
- Even if several attempts were made to obtain a linear function linking liquid thermal conductivity with temperature, the equations proposed up to now do not possess the generality of Eq. (4); moreover, Eq. (4) contains only one parameter characteristic of each compound.
- Equation (4) represent the saturated liquid thermal conductivity in the reduced temperature range from the reduced freezing point to well above 0.90.

The last step to be accomplished is concerned with the factor B , which can be linked with the critical pressure, the critical temperature, and the molecular mass: two forms for its estimation are found, each referred to the number of carbon atoms present in the fluid molecule, i.e., to the methane series of refrigerants and to the ethane series. The use of the normal boiling point T_b and of the normal freezing point T_f was avoided (even though satisfactory results were obtained with these easily available properties) because of their dependence on the normal pressure.

The result for the refrigerant series is

$$B = B^* T_c^\alpha P_c^\beta M^\gamma \quad (5)$$

where B^* , α , β , and γ are numerical constants whose values, different for the two series of methane and ethane, are reported in Table I.

TABEL I. Coefficients of Eq. (5)

Halocarbon refrigerants in the	B^*	α	β	γ
Methane series	0.4	1/4	1/3	-3/4
Ethane series	2.8	-1/6	1/6	-1/2

Table II. Comparison Between Experimental and Predicted Thermal Conductivities of Pure Refrigerants^a

Refrigerant	Formula	M	T _c (K)	P _c (kPa)	T _r expt. range	Number of expt. points	Source of expt. data	Constant B of Eq. (5)	AAD (%)	MAD (%)
R10	CCl ₄	153.82	556.30	44.13	0.45-0.85	23	[6]	0.157	9.6	11.6
R11	CCl ₃ F	137.37	471.20	44.06	0.49-0.93	25	[7-9]	0.164	1.2	3.8
R12	CCl ₂ F ₂	120.91	384.95	41.13	0.53-0.92	25	[7-9]	0.168	3.6	8.5
R13	CClF ₃	104.46	302.01	38.65	0.33-0.90	15	[6, 7]	0.173	2.8	6.9
R13B1	CBrF ₃	148.91	340.20	39.62	0.33-0.98	31	[6, 7]	0.137	5.8	9.7
R22	CHClF ₂	86.47	369.30	49.90	0.31-0.96	27	[6-9]	0.228	5.4	11.0
R23	CHF ₃	70.01	299.10	48.33	0.57-0.94	12	[10]	0.250	2.2	4.2
R113	CCl ₂ FCClF ₂	187.38	487.50	34.37	0.52-0.92	11	[7, 8, 11]	0.131	0.9	2.2
R114	CClF ₂ CClF ₂	170.92	418.80	32.48	0.46-0.92	27	[6-8]	0.140	3.9	9.8
R115	CClF ₂ CF ₃	154.47	353.05	31.53	0.48-0.97	28	[6, 7, 13]	0.151	8.4	11.2
R123	CHCl ₂ CF ₃	152.93	456.94	36.68	0.55-0.78	44	[8, 14-17]	0.149	1.5	3.7
R123a	CHClFCClF ₂	152.93	461.10	44.70	0.67-0.74	8	[8]	0.153	3.2	4.3
R124	CHClFCF ₃	136.47	395.62	36.34	0.59-0.93	22	[7, 8, 18]	0.161	1.4	4.7
R134a	CF ₃ CH ₂ F	102.03	374.30	40.60	0.59-0.97	96	[8, 14-17, 20-25]	0.191	6.6	10.5
R141b	CH ₃ CCl ₂ F	116.94	477.30	41.20	0.50-0.82	16	[18-23]	0.172	3.7	9.3
R142b	CH ₃ CClF ₂	100.49	410.30	41.20	0.61-0.97	18	[18-24]	0.190	7.1	9.6
R152a	CHF ₂ CH ₃	66.05	386.70	44.92	0.66-0.94	17	[16-18]	0.241	3.4	5.5

^a AAD(%) = $\text{abs}[(\sum (\lambda_{\text{calc}}/\lambda_{\text{exp}} - 1))/n] \cdot 100$; MAD(%) = $\text{max of } [\text{abs}(\lambda_{\text{calc}}/\lambda_{\text{exp}} - 1)] \cdot 100$; λ_{exp} = experimental thermal conductivity value; λ_{calc} = estimated thermal conductivity value; n = number of experimental points.

A first method's validation has been carried out of comparison with the experimental data used to implement Eqs. (4) and (5); most of the data are very recent, due to the huge effort spent on the study of thermophysical properties of alternative refrigerants. The validation results are summarized in Table II, where all the sources of experimental data are reported with

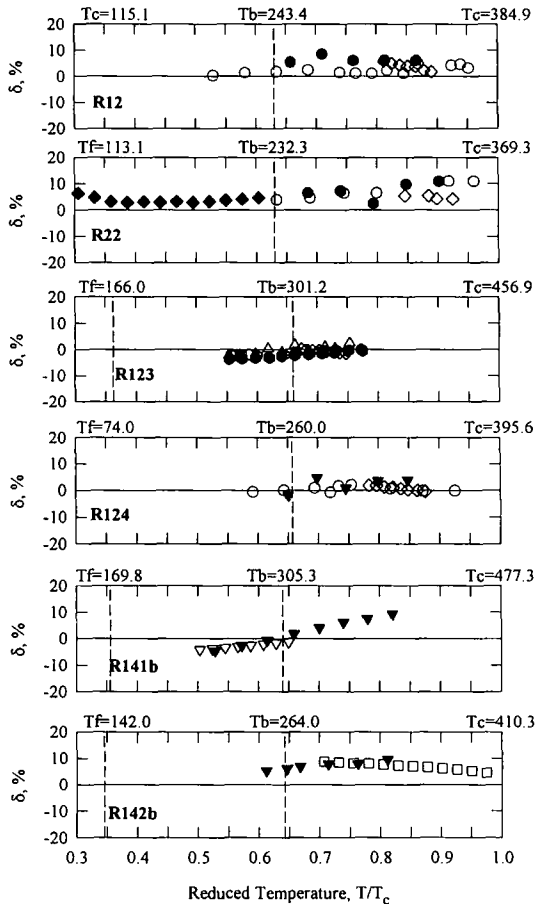


Fig. 1. Deviation between predicted thermal conductivities and experimental values for some CFC and HCFC refrigerants. $\delta = 100(\lambda_{\text{calc}}/\lambda_{\text{exp}} - 1)$. T_f , normal freezing temperature in K; T_b , normal boiling temperature in K; T_c , critical temperature in K. \blacklozenge [6]; \circ [7]; \diamond [8]; \bullet [9]; \triangle [14]; \circ [15]; \bullet [16]; \blacktriangle [17]; \diamond [18]; ∇ [23]; \square [24].

the average absolute deviation (AAD) and the maximum absolute deviation (MAD) parameters for each refrigerant.

The method gives average absolute deviations between estimated and experimental thermal conductivity usually less than 5% as can be seen from Table II. Figures 1 and 2 show the same information in more detail for each refrigerant and for each reference.

A more significant validation of the method's predictive power was then achieved by comparing experimental and calculated thermal conductivities of several refrigerants, namely, R20, R21, R32, R125, and R133a, whose experimental data were not used to implement the present correlation; the results reported in Table III and Fig. 2 show the same good agreement between estimated and predicted λ values also for these fluids.

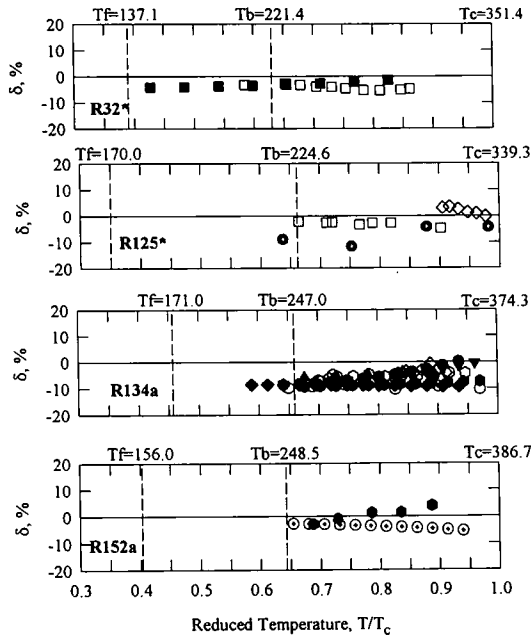


Fig. 2. Deviation between predicted thermal conductivities and experimental values for some HFC refrigerants. $\delta = 100(\lambda_{calc}/\lambda_{exp} - 1)$. T_f , normal freezing temperature in K; T_b , normal boiling temperature in K; T_c , critical temperature in K. (*) Refrigerant not used to implement the method. \diamond [8]; \blacksquare [11]; \square [12]; \triangle [14]; \circ [15]; \oplus [16]; \blacktriangle [17]; \bullet [18]; \oplus [19]; \circ [20]; \bullet [21]; \blacktriangledown [22]; ∇ [23]; \blacklozenge [25].

Table III. Comparison Between Experimental and Predicted Thermal Conductivities of Pure Refrigerants Not Used to Implement the Prediction Method^a

Refrigerant	Formula	M	T _c (K)	P _c (kPa)	T _r expt. range	Number of expt. points	Source of expt. data	Constant B of Eq. (5)	AAD (%)	MAD (%)
R20	CHCl ₃	119.38	536.40	53.84	0.50-0.63	8	[6]	0.201	1.8	3.5
R21	CHCl ₂ F	102.93	451.65	51.70	0.45-0.96	16	[6]	0.213	6.6	8.6
R32	CH ₂ F ₂	52.02	351.36	58.14	0.43-0.86	18	[11, 12]	0.346	3.8	5.4
R125	CHF ₂ CF ₂	120.03	339.33	36.39	0.64-0.98	17	[8, 12, 19]	0.176	3.6	11.7
R133a	CH ₂ ClCF ₃	118.49	432.00	49.40	0.39-0.72	8	[11]	0.179	4.4	6.4

^a AAD(%) = $\text{abs}\{[\sum(\dot{\lambda}_{\text{calc}}/\dot{\lambda}_{\text{exp}} - 1)]/n\}$ 100; MAD(%) = $\text{max of } [\text{abs}(\dot{\lambda}_{\text{calc}}/\dot{\lambda}_{\text{exp}} - 1)]100$; $\dot{\lambda}_{\text{exp}}$ = experimental thermal conductivity value; $\dot{\lambda}_{\text{calc}}$ = estimated thermal conductivity value; n = number of experimental points.

4. APPLICATION TO MIXTURES

As far as the application of the method to mixtures is concerned, a general correlation for binary organic liquid mixtures, based on a simple "quadratic mixing law," can be generalized to obtain the following equation [2, 4]:

$$\lambda = (B_1 x_1^2 + B_2 x_2^2 + 2.20 \sqrt{B_1^3 B_2} x_1 x_2) (1 - \frac{3}{4} T_r) \quad (6)$$

where x_1 and x_2 are the molar fractions of the two components, B_1 and B_2 are the coefficients B of Eq. (9), with $B_1 < B_2$; and T_r is calculated using a critical temperature evaluated with Kay's rule [3].

As very scanty experimental data exist, to the authors' knowledge, on thermal conductivity of mixtures, the check was developed for only one azeotropic mixture (R502) and for several nonazeotropic mixtures composed of R22, R142b, and R152a, as shown in Table IV. The experimental data for the latter mixtures [26] were obtained for the subcooled liquid at different pressures. The data used for comparison refer to saturation for the azeotropic mixture R502 and to the pressure nearest to saturation (namely, 2.1 MPa) for the other mixtures; as can be seen, the method shows similar deviations in both cases.

5. CONCLUSIONS

A prediction method, based upon an extensive analysis of the existing experimental data, is proposed for the thermal conductivity of halocarbon refrigerants in the liquid state along the saturation line and in the subcooled region at pressures near saturation: the method is validated against experimental data in the reduced temperature range from the reduced freezing point to well above 0.90.

The aim of the method is to allow very simple and handy calculation of the transport property with an accuracy better than 10%, which will be useful for engineering purposes. The method determines the thermal conductivity as a sole function of the reduced temperature, through the knowledge of a monomial parameter dependent on easily available physical constants characteristic of each substance, namely, the critical temperature, critical pressure, and molecular weight. The numerical constants to be used in the monomial parameter are given for both the methane and the ethane series of the halocarbon refrigerants.

The method validation, carried out also with data of refrigerants not used to obtain Eqs. (4) and (5), shows average absolute deviations which are generally less than 5%, with maximum absolute deviations usually less than 10% in the reduced temperature range 0.3 to well above 0.9.

Table IV. Comparison Between Experimental and Predicted Thermal Conductivities of Refrigerant Mixtures^a

Mixture	Comp. 1	% weight	Comp. 2	% weight	T_r expt. range	N of expt. points	Source of expt. data	B estimated from Eq. (6)	AAD (%)	MAD (%)
Azeotropic R502	R22	0.488	R115	0.512	0.85–0.90	5	[8]	0.157	3.0	3.6
Nonazeotropic R22/R142b ^b	R22	0.2796	R142b	0.7204	0.56–0.81	5	[26]	0.195	5.3	6.6
	R22	0.4350	R142b	0.5650	0.57–0.83	5	[26]	0.199	5.4	7.3
	R22	0.7290	R142b	0.2710	0.59–0.85	5	[26]	0.212	6.2	8.3
	R22	0.2488	R152a	0.7512	0.58–0.84	5	[26]	0.241	3.4	3.9
R22/R152a ^b	R22	0.4968	R152a	0.5032	0.59–0.85	5	[26]	0.240	5.2	7.0
	R22	0.7505	R152a	0.2495	0.60–0.86	5	[26]	0.236	7.8	9.5

^a AAD(%) = $\text{abs}\{[\sum(\lambda_{\text{calc}}/\lambda_{\text{exp}} - 1)]/n\} \cdot 100$; MAD(%) = $\text{max}\{[\text{abs}(\lambda_{\text{calc}}/\lambda_{\text{exp}} - 1)]/100\}$; λ_{exp} = experimental thermal conductivity value;

λ_{calc} = estimated thermal conductivity value; n = number of experimental points.

^b Experimental data obtained at 2.1 MPa.

An extension of the method to estimate the thermal conductivity of binary mixtures is also presented, along with comparisons with the experimental data available in literature. AAD and MAD are in this case generally less than 6 and 10%, respectively.

The method proposed for the refrigerant fluids can be extended to organic compounds and the results will be presented in a future paper.

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