International Journal of Thermophysics, Vol. 26, No. 5, September 2005 (© 2005)
DOI: 10.1007/s10765-005-8095-4

Equations for the Thermal Conductivity of R-32, R-125, R-134a, and R-143a[1]

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Received May 19, 2005

At present hydrofluorocarbons (HFCs) such as R32, R-125, R-134a, and R-143a are widely used, and it is required to obtain accurate information of thermophysical properties, especially of the thermal conductivity of HFCs. In this paper new thermal conductivity equations for R-32, R-125, R134a, and R143a are proposed, applicable over a wide range of temperature and pressure including the critical region based on existing experimental data, and the reliability of the present equations is summarized. The problem that the thermal conductivity calculated from the thermal diffusivity in the critical region differs depending on the equation of state is also discussed.

KEY WORDS: correlating equation; HFCs; R-32; R-125; R-134a; R-143a; thermal conductivity.

1. INTRODUCTION

Although the global warming potentials (GWPs) of hydrofluorocarbons (HFCs) such as R-32 (difluoromethane), R-125 (pentafluoroethane), R134a (1,1,1,2-tetrafluoroethane), and R143a (1,1,1-trifluoroethane) are relatively large, they are widely used at present. It will take time to replace HFCs by more preferable substances such as natural refrigerants. Thus, it is necessary to improve the present knowledge of thermophysical properties, especially of the transport properties of HFCs.

[1] Paper presented at the Sixteenth European Conference for Thermophysical Properties, September 1–4, 2002, London, United Kingdom.

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Up to now, we have an equation for the thermal conductivity of R-134a by Krauss et al. [1] and ones for R-32, R-125, and R-134a by Kiselev et al. [2]. These equations are applicable over a wide range including the critical region. However, recently important new sets of data for the thermal conductivity have been reported and it is necessary to formulate new thermal conductivity equations of HFCs that include these new sets of data.

Following a previous paper [3] on natural refrigerants, in this paper new equations for the thermal conductivity of R-32, R-125, R134a, and R-143a for practical use, applicable over a wide range of temperature and pressure, are proposed based on the available experimental data.

2. AVAILABLE EXPERIMENTAL DATA

Data sets of the thermal conductivity of R-32 [4–14], R-125 [4, 7, 8, 10, 12–23], R-134a [7, 11, 15, 24–33], and R-143a [13, 14, 30, 34, 35] considered in the present formulation are listed in Tables I to IV. Experimental data for R-32 are also shown in Fig. 1. Recently extensive data sets at high temperatures and pressures have been reported by Le Neindre and his coworkers for these substances [9, 19, 28, 35]. In the critical region few thermal conductivity data have been reported. However, thermal diffusivity data have been reported by Kruppa and Straub [33] for R-134a, by Pitschmann and Straub [14] for R-32, R125, and R-143a, and thermal conductivity values in the critical region can be calculated from thermal diffusivity data by using appropriate equations of state.

3. EQUATIONS

The equation for the thermal conductivity of HFCs is expressed as

$$\lambda(T, \rho) = \lambda_0(T) + \lambda_1(\rho) + \Delta\lambda_c(T, \rho) \tag{1}$$

where λ_0 denotes the ideal gas thermal conductivity and is a function of temperature only. $\lambda - \lambda_0$ denotes the excess thermal conductivity and at high temperatures it reduces to $\lambda_1(\rho)$ which is a function of density only. $\Delta\lambda_c$ denotes the critical enhancement of thermal conductivity and has effective values only in the critical region.

The term λ_0 is expressed as follows:

$$\lambda_0(T) = a_0 + a_1(T/T_c) + a_2(T/T_c)^2, \tag{2}$$

and the term λ_1 is assumed as follows:

$$\lambda_1(\rho) = b_1(\rho/\rho_c) + b_2(\rho/\rho_c)^2 + b_3(\rho/\rho_c)^3 + b_4(\rho/\rho_c)^4 + b_5(\rho/\rho_c)^5 \tag{3}$$

Table I. Experimental Data for the Thermal Conductivity of R-32[a]

Author (1st)	Ref. No.	Year	Method	Purity (mol%)	T (K)	P (MPa)	N	State
R-32								
Assael	4	1995	THW	99.98	253–334	Sat–20	27	L
Gao	5	1995	THW	99.98	193–303	0.2–30	44	L
Geller	6	1994	SHW	–	253–427	0.1–15	122	LVC
Grebenkov	7	1994	SCC	99.99	275–400	1.7–13	95	L
Gross	8	1996	THW	99.8	233–363	0.1–6	80	LV
Le Neindre	9	2001	SCC	99.9	300–465	0.1–50	582	LV
Papadaki	10	1993	THW	99.9	205–303	Sat	10	L
Ro	11	1995	THW	99.9	223–323	2–20	24	L
Tanaka	12	1995	THW	99.9	283–333	0.1–Sat	53	V
Yata	13	1996	THW	99.5	253–324	Sat–30	27	L
Pitschmann[b]	14	2000	DLS	99.9	300–390	1–10	210	C

[a] Ref. No.: Reference number; N: Number of data; THW: Transient hot-wire method; TCC: Transient coaxial-cylinder method; SHW: Steady-state hot-wire method; SCC: Steady-state coaxial-cylinder method; DLS: Dynamic light-scattering method; Sat: Saturated state; L: Liquid phase; V: Vapor phase; C: Critical region.
[b] Thermal diffusivity data.

Table II. Experimental Data Sets for the Thermal Conductivity of R-125 (Symbols are the same as in Table I)

Author (1st)	Ref. No.	Year	Method	Purity (mol%)	T (K)	P (MPa)	N	State
R-125								
Assael	15	1995	THW	99.95	273–333	0.1–Sat	5	V
Assael	4	1995	THW	99.95	253–334	Sat–20	20	L
Fellows	16	1990	THW	99.8	283–343	Sat	6	LV
Gao	17	1995	THW	99.6	193–333	1–30	32	L
Grebenkov	7	1994	SCC	99.90	290–405	0.2–20	74	L
Gross	8	1996	THW	99.8	233–363	0.1–6	78	LV
Kim	18	1995	THW	99.8	223–323	2–20	24	L
Le Neindre	19	1999	SCC	99.8	300–515	0.1–53	610	LV
Sun	20	1997	THW	99.95	251–334	Sat	17	V
Papadaki	10	1993	THW	99.9	225–306	Sat	7	L
Tanaka	12	1995	THW	99.8	283–333	0.1–Sat	51	V
Tsvetkov	21	1993	TCC	99.83	160–293	0.1–10	16	L
Tsvetkov	22	1995	TCC	99.83	187–419	0.1–6	30	LV
Wilson	23	1992	THW	99.7	216–333	Sat	7	LV
Yata	13	1996	THW	99.5	257–305	Sat–30	24	L
Pitschmann[a]	14	2000	DLS	99.9	303–373	1–9	256	C

[a] Thermal diffusivity data.

Table III. Experimental Data for the Thermal Conductivity of R-134a (Symbols are the same as in Table I)

Authors (1st)	Ref. No.	Year	Method	Purity (mol%)	T (K)	P (MPa)	N	State
R-134a								
Assael	24	1992	THW	99.9	253–343	Sat–23	36	L
Assael	15	1995	THW	99.95	273–333	0.1–Sat	7	V
Grebenkov	7	1994	THW	99.91	290–405	0.2–20	65	L
Gross	25	1992	THW	99.9	253–363	0.01–6	87	LV
Gurova	26	1996	THW	99.9	213–293	Sat–20	53	L
Leasecke	27	1992	THW	–	200–390	0.05–70	215	LVC
Le Neindre	28	1999	SCC	99.9	300–530	0.1–530	511	LV
Papadaki	29	1993	THW	99.9	240–307	Sat	9	L
Ro	11	1995	THW	99.8	223–323	2–20	24	L
Tanaka	30	1991	SCC	99.9	293–353	0.1–3	33	V
Yamamoto	31	1992	THW	99.9	273–363	0.01–3	38	V
Yata	32	1989	THW	99.5	253–333	0.1–30	21	L
Kruppa[a]	33	1992	DLS	–	290–420	0.8–8.3	127	C

[a] Thermal diffusivity data.

Table IV. Experimental Data for the Thermal Conductivity of R-143a (Symbols are the same as in Table I)

Authors (1st)	Ref. No.	Year	Method	Purity (mol%)	T (K)	P (MPa)	N	State
R-143a								
Lee	34	2000	THW	–	233–323	2–20	24	L
Le Neindre	35	2001	SCC	99.5	300–500	0.1–50	592	LV
Tanaka	30	1991	CC	99.9	293–353	0.1–4	30	V
Yata	13	1996	THW	99.5	268–314	Sat–30	24	L
Pitschmann[a]	14	2000	DLS	99.9	316–380	2–9	191	C

[a] Thermal diffusivity data.

In order to express the critical enhancement of the thermal conductivity, the term $\Delta\lambda_c$ is necessary. In this study the approximate treatment by Olchowy and Sengers [36] is adopted. They give an approximate equation for $\Delta\lambda_c$ as follows:

$$\Delta\lambda_C(T, \rho) = \rho C_P \frac{R_0 kT}{6\pi\eta\xi}(\tilde{\Omega} - \tilde{\Omega}_0) \tag{4}$$

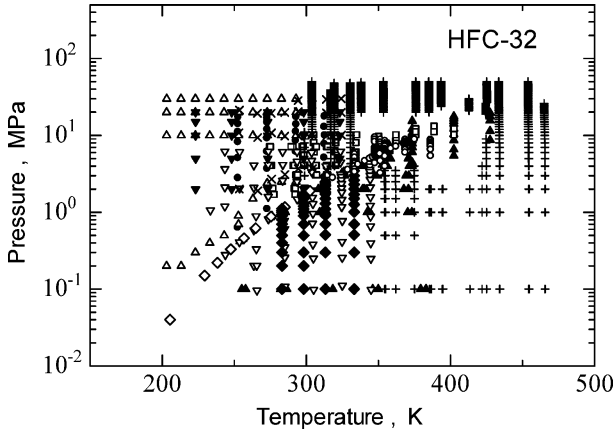

Fig. 1. Experimental data for thermal conductivity of R-32: (●) Assael and Karagiannidis [4]; (△) Gao et al. [5]; (▲) Geller and Paulaitis [6]; (□) Grebenkov et al. [7]; (▽) Gross and Song [8]; (+) Le Neindre and Garrabos [9]; (◇) Papadaki and Wakeham [10]; (▼) Ro et al. [11]; (⧫) Tanaka et al. [12]; (×) Yata et al. [13]; (◯) Pitschmann and Straub [14].

where

$$\tilde{\Omega} = \frac{2}{\pi}\left[\left(\frac{C_p - C_v}{C_p}\right)\tan^{-1}(\bar{q}_D\xi) + \frac{C_v}{C_p}\bar{q}_D\xi\right] \tag{5}$$

$$\tilde{\Omega}_0 = \frac{2}{\pi}\left\{1 - \exp\left[\frac{-1}{(\bar{q}_D\xi)^{-1} + (1/3)(\bar{q}_D\xi\rho_c/\rho)^2}\right]\right\} \tag{6}$$

$$\xi = \xi_0(\Delta\tilde{\chi}/\Gamma)^{v/\gamma} \tag{7}$$

$$\Delta\tilde{\chi} = \tilde{\chi}(T, \rho) - \tilde{\chi}(T_R, \rho)T_R/T \tag{8}$$

$$\tilde{\chi}(T, \rho) = \frac{P_c\rho}{\rho_c^2}\left(\frac{\partial\rho}{\partial P}\right)_T \tag{9}$$

In the equations above C_p, C_v, and η denote the specific heat capacity at constant pressure, the specific heat capacity at constant volume, and the viscosity, respectively. T_R is the reference temperature ($=2.0T_c$) and k is the Boltzmann constant. $R_0(=1.01)$ is a system-independent universal constant and $v(=0.63)$ and $\gamma(=1.239)$ are the system-independent universal exponents. Γ and ξ_0 are the system-dependent constants and are determined from the equation of state for each substance. The cutoff parameter $\bar{q}_D$ for each substance is determined by using the experimental data of the thermal conductivity in the critical region.

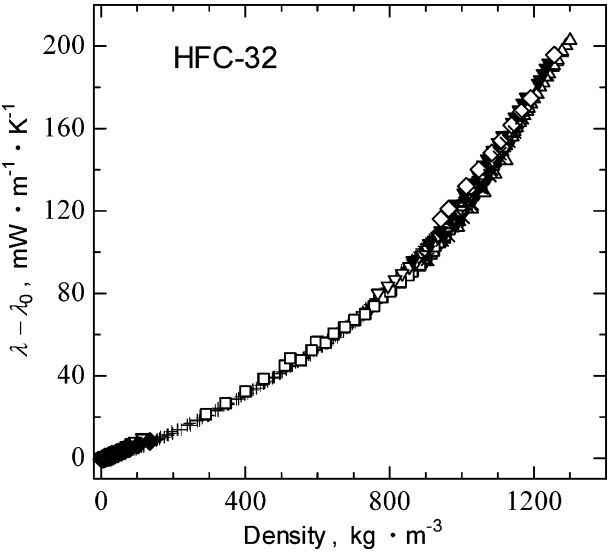

Fig. 2. Excess thermal conductivity of R-32 outside the critical region as a function of density: (●) Assael and Karagiannidis [4]; (△) Gao et al. [5]; (□) Grebenkov et al. [7]; (▽) Gross and Song [8]; (+) Le Neindre and Garrabos [9]; (◇) Papadaki and Wakeham [10];(▼) Ro et al. [11]; (⧫) Tanaka et al. [12]; (×) Yata et al. [13].

4. DETERMINATION OF EQUATIONS OUTSIDE CRITICAL REGION

Outside the critical region, the thermal conductivity can be expressed by the terms λ_0 and λ_1. First of all, the values of coefficients for the four HFCs in Eq. (2) are determined by using data in the gaseous state near atmospheric pressure. Figure 2 shows the relation between $\lambda - \lambda_0$ and density ρ for R-32 excluding data in the critical region. Outside the critical region, $\lambda - \lambda_0$ can be expressed as a function of density only, and in this region the coefficients $\lambda_1 = \lambda - \lambda_0$ in Eq. (3) for the four HFCs are determined. Figure 3 shows deviations of the same data in Fig. 2 for R-32. Numerical values of the coefficients in Eqs. (2) and (3) are tabulated in Table V.

5. THERMAL CONDUCTIVITY IN THE CRITICAL REGION

In the critical region direct measurements of the thermal conductivity were only reported by Geller et al. [6] for R-32. However, Kruppa and Straub [33] and Pitschmann and Straub [14] reported thermal diffusivity

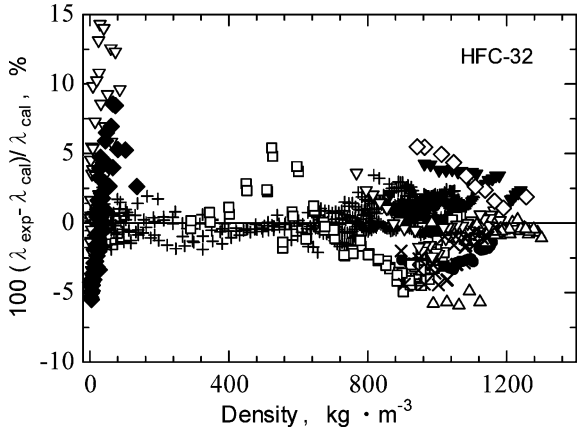

Fig. 3. Deviations of experimental data for thermal conductivity of R-32 from the equation outside the critical region: (●) Assael and Karagiannidis [4]; (△) Gao et al. [5]; (□) Grebenkov et al. [7]; (▽) Gross and Song [8]; (+) Le Neindre and Garrabos [9]; (◇) Papadaki and Wakeham [10]; (▼) Ro et al. [11]; (◆) Tanaka et al. [12]; (×) Yata et al. [13].

Table V. Numerical Values of Coefficients in Equations for the Thermal Conductivity of HFCs

	R-32	R-125	R-134a	R-143a
T_C(K)	351.26	339.17	374.27	345.97
P_C(MPa)	5.777	3.618	4.065	3.769
ρ_C(kg·m^{-3})	424.0	568.0	511.0	429.0
a_0(mW·m^{-1}·K^{-1})	−2.1095	−5.1855	−6.3643	−18.606
a_1(mW·m^{-1}·K^{-1})	6.6708	18.413	19.099	37.098
a_2(mW·m^{-1}·K^{-1})	12.485	4.1858	6.8704	0.0000
b_1(mW·m^{-1}·K^{-1})	13.232	2.8010	4.6754	13.843
b_2(mW·m^{-1}·K^{-1})	41.377	23.829	23.972	−5.0914
b_3(mW·m^{-1}·K^{-1})	−32.261	−20.162	−20.309	15.795
b_4(mW·m^{-1}·K^{-1})	13.199	8.3393	8.4665	−9.1248
b_5(mW·m^{-1}·K^{-1})	−1.7047	−0.99375	−0.98732	1.8742
Γ	0.057	0.052	0.051	0.054
ξ_0(nm)	0.164	0.187	0.185	0.188
$(\bar{q}_D)^{-1}$(nm)	1.44	1.86	0.91	1.15

data for HFCs in the critical region. The thermal conductivity can be calculated from the thermal diffusivity α by using the equation,

$$\lambda = \alpha \rho c_p \tag{10}$$

In the conversion from the thermal diffusivity to the thermal conductivity, values of density and isobaric heat capacity are needed. In the critical region, these values differ significantly depending on the equation of state used, which means the values of the thermal conductivity differ depending on the equation of state used. After reviewing the existing equations of state, the crossover equation of state by Kiselev and Huber [37] for R-32, R-125, and R-134a was found to be the most preferable. As the applicable range of the equation of state is limited to the critical region, it is only used for the purpose of conversion. And for the calculation of the thermal conductivity over the entire range including the critical region by using Eqs. (1) to (9), the equation of state by Tillner-Roth and Yokozeki [38] for R-32, that by Sunaga et al. [39] for R-125, that by Tillner-Roth and Baehr [40] for R-134a, and that by Outcalt and McLinden [41] for R-143a were used. By using the equations of state mentioned above and the values of the thermal conductivity calculated from the thermal diffusivity, the term expressed by Eq. (4) was determined for the four HFCs, and the values of the coefficients are tabulated in Table V. In the correlations of the equations and calculations of the thermal conductivity, the required thermophysical properties are calculated by using REFPROP [42].

Figures 4 and 5 show comparisons between the calculated and experimental values of critical enhancement of the thermal conductivity for R-32. In Fig. 4 the thermal conductivity is calculated with the equation of state by Tillner-Roth and Yokozeki [38], while in Fig. 5 it is calculated with the crossover equation of state by Kiselev and Huber [37]. The behavior of the thermal conductivity in the critical region for R-32 seems to be improved by using the crossover equation of state in the conversion from the thermal diffusivity. The same is true for R-125 and R-134a.

6. RELIABILITY OF THE PRESENT EQUATIONS

The experimental data are compared with the equations, and the average deviation of each set of data from the equation for each substance is tabulated in Tables VI to IX. The deviations of data sets converted from thermal diffusivity values are not included in the tables, because the conversion was not successful at some states. From detailed comparisons with the experimental data, the uncertainty of the present equations for

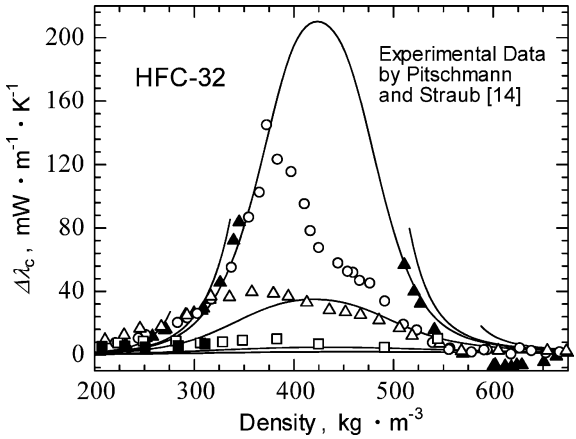

Fig. 4. Comparison of critical enhancement of thermal conductivity calculated from thermal diffusivity data using EOS by Tillner-Roth and Yokozeki with Eq. (4) by Olchowy and Sengers (solid curves): (▲) 350.90 K; (◯) 351.60 K; (△) 354.76 K; (□) 368.81 K; (■) 386.37 K.

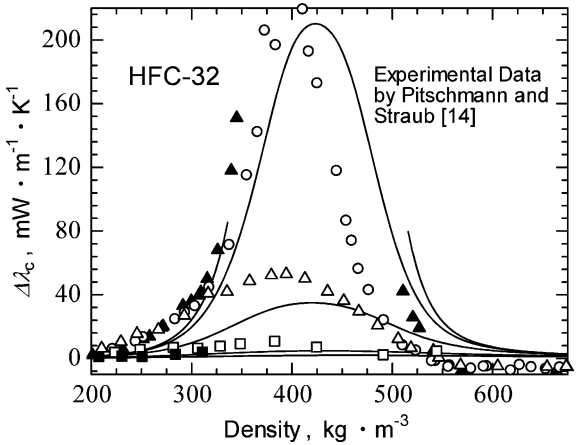

Fig. 5. Comparison of critical enhancement of thermal conductivity calculated from thermal diffusivity data using EOS by Kiselev and Huber with Eq. (4) by Olchowy and Sengers (solid curves): (▲) 350.90 K; (◯)351.60 K; (△) 354.76 K; (□) 368.81 K; (■) 386.37 K.

Table VI. Average Deviations of Experimental Data for R-32 from the Equation

Author (1st)	Ref. No.	Year	Dev. (%)
Assael	4	1995	2.6
Gao	5	1995	1.2
Geller	6	1994	11.1
Grebenkov	7	1994	2.4
Gross	8	1996	4.1
Le Neindre	9	2001	0.9
Papadaki	10	1993	3.3
Ro	11	1995	2.9
Tanaka	12	1995	3.5
Yata	13	1996	2.4
Pitschmann[a]	14	2000	–

[a] Thermal diffusivity data.

Table VII. Average Deviations of Experimental Data for R-125 from the Equation

Author (1st)	Ref. No.	Year	Dev. (%)
Assael	15	1995	3.9
Assael	4	1995	0.9
Fellows	16	1990	3.8
Gao	17	1995	0.7
Grebenkov	7	1994	3.8
Gross	8	1996	8.4
Kim	18	1995	1.6
Le Neindre	19	1999	0.7
Sun	20	1997	4.6
Papadaki	10	1993	2.0
Tanaka	12	1995	3.6
Tsvetkov	21	1993	3.4
Tsvetkov	22	1995	2.8
Wilson	23	1992	5.9
Yata	13	1996	1.7
Pitschmann[a]	14	2000	–

[a] Thermal diffusivity data.

the four HFCs in the temperature range from 200 to 600 K and in the pressure range up to 50 MPa is estimated to be ±2 to 3% in the liquid phase, ±3 to 5% in the gaseous phase, and ±5 to 15% in the critical region.

Table VIII. Average Deviations of Experimental Data for R-134a from the Equation

Author (1st)	Ref. No.	Year	Dev. (%)
Assael	24	1992	0.5
Assael	15	1995	4.6
Grebenkov	7	1994	1.8
Gross	25	1992	5.1
Gurova	26	1996	1.2
Laesecke	27	1992	3.6
Le Neindre	28	1999	0.8
Papadaki	29	1993	1.9
Ro	11	1995	0.5
Tanaka	30	1991	3.9
Yamamoto	31	1992	2.8
Yata	32	1989	0.5
Kruppa[a]	33	1992	–

[a] Thermal diffusivity data.

Table IX. Average Deviations of Experimental Data for R-143a from the Equation

Author (1st)	Ref. No.	Year	Dev. (%)
Lee	34	2000	3.0
Le Neindre	35	2001	1.1
Tanaka	30	1991	9.1
Yata	13	1996	2.7
Pitschmann[a]	14	2000	–

[a] Thermal diffusivity data.

7. CONCLUSIONS

Equations for the thermal conductivity of HFCs, namely R-32, R-125, R-134a, and R-143a applicable for practical use over a wide range of temperature and pressure including the critical region are proposed based on the experimental data published up to now. The uncertainty of the present equations in the temperature range from 200 to 600 K and in the pressure range up to 50 MPa is estimated.

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