International Journal of Thermophysics, Vol. 9, No. 4, 1988

Thermal Conductivity of Liquid Halogenated Ethanes Under High Pressures

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Received June 2, 1988

New experimental data on the thermal conductivity of liquid halogenated ethanes, R112 (CCl_2F-CCl_2F), R113 ($CCl_2F-CClF_2$), R114 ($CClF_2-CClF_2$), R114B2 ($CBrF_2-CBrF_2$), and R123 ($CHCl_2-CF_3$), are presented in the temperature range from 283 to 348 K at pressures up to 200 MPa or the freezing pressures. The measurements were carried out by a transient hot-wire apparatus within an uncertainty of $\pm 1.0\%$. The thermal conductivity data obtained have been analyzed by means of the corresponding-states principle and other empirical methods. It is found that the corresponding-states correlation $\lambda = f(T_r, P_r)$ holds well for R112, R113, and R114. The thermal conductivity can also be correlated satisfactorily with temperature, pressure, and molar volume by a similar expression to the Tait equation and the dense hard-sphere model presented by Dymond.

KEY WORDS: fluorocarbon; halogenated ethane; liquid; high pressure; refrigerant; thermal conductivity; transient hot-wire method.

1. INTRODUCTION

Fluorocarbons and their mixtures have been used extensively in refrigeration, air conditioning, plastic foam blowing, cleaning, and fire-extinguishing applications. They have been also considered as promising working fluids in various types of power plants. However, serious problems have arisen recently for the release of fluorocarbons in connection with the new environmental consequences, such as the greenhouse effect in the troposphere and the ozone depletion in the stratosphere. It is, therefore, an urgent need to search appropriate alternatives to fluorocarbons employed

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0195-928X/88/0700-0465$06.00/0

840/9/4-1

so far. The accumulation of reliable numerical data and knowledge on the thermophysical properties of these fluids is also desired.

The present investigation was undertaken in order to obtain accurate thermal conductivity data of liquid halogenated ethanes, R112, R113, R114, R114B2, and R123, in the temperature range from 283 to 348 K at pressures up to 200 MPa or the freezing pressures. Among them, R123 is a new compound which is considered as a candidate of alternatives to R11 (CCl_3F).

2. EXPERIMENTAL

The measurements have been performed on an absolute basis by a transient hot-wire apparatus fully described elsewhere [1] with an uncertainty of less than 1.0%. The operating procedures and the method of data processing also remained unchanged. The thermal conductivity under a given condition of temperature and pressure was reproducible within 0.1%. The hot-wire cell was thermostatically controlled to within 10 mK. The pressure was measured by the calibrated Bourdon-tube gauges within an error of 0.1%. The purified sample fluids were supplied by Daikin Industries, Ltd., with purities better than 99.9%. Each of the experimental measurements has been subjected to the analysis proposed by Nieto de Castro et al. [2] in order to determine whether there is a significant radiative contribution to the thermal conductivity. After the analysis it is concluded that the experimental results reported here are radiation-free values.

3. EXPERIMENTAL RESULTS

Prior to measuring the thermal conductivity of liquid fluorocarbons, that of toluene was measured. Toluene has been often used as a calibration fluid for the measurement of the thermal conductivity of liquids. It was confirmed that the present results agreed with the previous work by Kashiwagi et al. [1] within 0.7%.

The experimental results for fluorocarbons investigated are summarized in Tables I to V. Each table contains the thermal conductivity at the reference state and that adjusted to nominal temperatures. A small temperature correction was made by means of the following equation:

$$\lambda(P, T_{\text{nom}}) = (P, T_{\text{ref}}) + (\partial\lambda/\partial T)_{P,T_{\text{nom}}}(T_{\text{nom}} - T_{\text{ref}}) \tag{1}$$

The derivative $(\partial\lambda/\partial T)_{P,T_{\text{nom}}}$ was evaluated with the assumption that the thermal conductivity of the liquids is a linear function of temperature at a constant pressure.

Table I. Thermal Conductivity of R112

T_{nom} (K)	T_{ref} (K)	P (MPa)	λ_{ref} ($W \cdot m^{-1} \cdot K^{-1}$)	λ_{nom} ($W \cdot m^{-1} \cdot K^{-1}$)
303.15	303.91	0.10	0.0779	0.0780
313.15	313.91	0.10	0.0759	0.0760
	313.82	20.10	0.0819	0.0820
323.15	323.81	0.10	0.0740	0.0741
	323.76	25.06	0.0803	0.0804
	323.88	35.95	0.0831	0.0832
338.15	338.77	0.10	0.0712	0.0713
	338.71	25.06	0.0787	0.0788
	338.72	50.02	0.0843	0.0844
	338.74	57.67	0.0863	0.0864
348.15	348.78	0.10	0.0694	0.0695
	348.77	25.06	0.0767	0.0768
	348.74	50.02	0.0835	0.0836
	348.74	72.50	0.0871	0.0872

Table II. Thermal Conductivity of R113

T_{nom} (K)	T_{ref} (K)	P (MPa)	λ_{ref} ($W \cdot m^{-1} \cdot K^{-1}$)	λ_{nom} ($W \cdot m^{-1} \cdot K^{-1}$)
298.15	298.86	0.10	0.0709	0.0710
	298.85	24.99	0.0782	0.0783
	298.76	50.02	0.0849	0.0850
	298.85	75.05	0.0894	0.0895
	298.79	100.01	0.0943	0.0944
	298.80	125.03	0.0991	0.0992
313.15	313.86	0.10	0.0678	0.0679
	313.81	25.06	0.0748	0.0749
	313.78	50.02	0.0815	0.0816
	313.88	75.05	0.0868	0.0869
	313.85	100.01	0.0923	0.0924
	313.84	125.03	0.0968	0.0969
	313.80	149.99	0.1017	0.1018
323.15	323.91	1.48	0.0659	0.0660
	323.81	25.06	0.0730	0.0731
	323.76	50.02	0.0801	0.0802
	323.76	75.05	0.0856	0.0857
	323.77	100.01	0.0904	0.0905
	323.76	124.90	0.0955	0.0956
	323.72	149.99	0.1001	0.1002
	323.70	175.02	0.1048	0.1049

3.1. Temperature Dependence of Thermal Conductivity

The temperature dependence of the thermal conductivity of R113 at atmospheric pressure is typically illustrated in Fig. 1 together with several literature values including some data at saturated pressures. The thermal conductivity of liquid R113 decreases almost linearly with rising temperature. It should be noted that there exist remarkable scatters among literature values. The earlier measurements before the development of the

Table III. Thermal Conductivity of R114

T_{nom} (K)	T_{ref} (K)	P (MPa)	λ_{ref} ($W \cdot m^{-1} \cdot K^{-1}$)	λ_{nom} ($W \cdot m^{-1} \cdot K^{-1}$)
283.15	283.77	25.01	0.0721	0.0722
	283.70	50.02	0.0796	0.0797
	283.78	75.05	0.0857	0.0858
	283.72	100.01	0.0914	0.0915
	283.67	125.03	0.0970	0.0971
	283.70	149.99	0.1016	0.1017
	283.71	175.02	0.1057	0.1058
	283.68	199.98	0.1094	0.1095
298.15	298.78	24.99	0.0686	0.0687
	298.88	50.02	0.0760	0.0762
	298.70	74.98	0.0826	0.0827
	298.62	100.01	0.0886	0.0887
	298.73	125.03	0.0943	0.0944
	298.81	149.99	0.0991	0.0992
	298.71	175.02	0.1032	0.1033
	298.62	200.05	0.1075	0.1076
313.15	313.61	24.99	0.0653	0.0654
	313.65	50.02	0.0730	0.0731
	313.62	74.98	0.0795	0.0796
	313.80	100.01	0.0860	0.0861
	313.74	125.03	0.0919	0.0920
	313.73	149.99	0.0969	0.0970
	313.84	175.02	0.1012	0.1013
	313.78	200.05	0.1053	0.1054
323.15	323.56	25.06	0.0626	0.0627
	323.47	50.02	0.0701	0.0702
	323.63	75.05	0.0769	0.0770
	323.61	100.01	0.0844	0.0845
	323.85	125.03	0.0901	0.0902
	323.71	150.06	0.0952	0.0953
	323.82	175.02	0.0995	0.0996
	323.66	199.98	0.1039	0.1040

transient hot-wire technique gave comparatively higher values. The data of Markwood and Benning [3] are higher than the present results by more than 20%. The recommended values of TPRC/CINDAS [10], which were generated based on several early data, are also 5% higher than ours. However, the present results, including the temperature coefficient of the thermal conductivity $(\partial\lambda/\partial T)_P$, agree satisfactorily with recent data of Yata et al. [12] and Cerneewa [4].

The isobaric temperature dependences of the thermal conductivity at 25 MPa for the fluorocarbons studied are plotted in Fig. 2. As for R112, R113, and R114, in which hydrogen atoms are completely substituted by chlorine or fluorine atoms, the thermal conductivity decreases and the absolute value of $(\partial\lambda/\partial T)_P$ increases with increasing number of fluorine

Table IV. Thermal Conductivity of R114B2

T_{nom} (K)	T_{ref} (K)	P (MPa)	λ_{ref} ($W \cdot m^{-1} \cdot K^{-1}$)	λ_{nom} ($W \cdot m^{-1} \cdot K^{-1}$)
298.15	298.82	0.10	0.0597	0.0598
	298.81	25.00	0.0653	0.0654
	298.90	50.01	0.0710	0.0711
	298.94	75.00	0.0753	0.0754
	298.88	100.01	0.0793	0.0794
	298.77	125.03	0.0831	0.0832
	298.87	149.99	0.0863	0.0864
	298.87	175.02	0.0891	0.0892
	298.94	200.05	0.0922	0.0923
313.15	313.96	0.10	0.0572	0.0573
	313.81	25.06	0.0627	0.0628
	313.84	50.02	0.0682	0.0683
	313.76	75.02	0.0728	0.0729
	313.79	100.01	0.0770	0.0771
	313.78	125.03	0.0813	0.0814
	313.84	149.99	0.0851	0.0852
	313.71	175.02	0.0876	0.0877
	313.65	200.05	0.0909	0.0910
323.15	324.01	2.17	0.0556	0.0557
	323.67	25.06	0.0605	0.0606
	323.61	50.02	0.0664	0.0665
	323.63	75.05	0.0711	0.0712
	323.61	100.01	0.0756	0.0757
	323.76	125.03	0.0799	0.0800
	323.73	149.99	0.0836	0.0837
	323.74	175.02	0.0866	0.0867
	323.73	200.05	0.0900	0.0901

atoms, respectively, as pointed out by Yata et al. [12] and Tauscher [9] including R115 and R116. The thermal conductivity of R114B2, which contains two heavy bromine atoms, is relatively low and that of R123, which contains one hydrogen atom, seems to be high compared with those of other ethane series of fluorocarbons.

Table V. Thermal Conductivity of R123

T_{nom} (K)	T_{ref} (K)	P (MPa)	λ_{ref} ($W \cdot m^{-1} \cdot K^{-1}$)	λ_{nom} ($W \cdot m^{-1} \cdot K^{-1}$)
283.15	283.66	0.10	0.0792	0.0793
	283.81	25.06	0.0888	0.0889
	283.76	50.02	0.0972	0.0973
	283.77	75.05	0.1030	0.1031
	283.73	100.01	0.1090	0.1091
	283.77	125.03	0.1144	0.1145
	283.80	149.99	0.1191	0.1192
	283.81	175.02	0.1241	0.1242
	283.82	199.98	0.1278	0.1279
298.15	298.79	6.31	0.0781	0.0782
	298.93	25.06	0.0858	0.0859
	298.82	50.02	0.0935	0.0937
	298.75	75.05	0.0998	0.0999
	298.90	100.01	0.1066	0.1067
	298.65	125.03	0.1121	0.1122
	298.77	150.03	0.1170	0.1171
	298.73	175.02	0.1216	0.1217
	298.70	200.05	0.1265	0.1266
313.15	313.74	2.51	0.0732	0.0733
	313.81	25.06	0.0844	0.0845
	313.80	50.02	0.0900	0.0902
	313.80	75.05	0.0975	0.0976
	313.74	100.01	0.1042	0.1043
	313.71	125.03	0.1093	0.1094
	313.73	149.99	0.1151	0.1152
	313.76	175.02	0.1194	0.1195
	313.72	199.98	0.1241	0.1242
323.15	323.78	8.03	0.0719	0.0720
	323.79	25.06	0.0808	0.0809
	323.65	50.02	0.0876	0.0878
	323.68	75.05	0.0956	0.0957
	323.69	100.01	0.1019	0.1020
	323.66	125.03	0.1077	0.1078
	323.71	149.99	0.1131	0.1132
	323.89	175.02	0.1177	0.1178
	323.83	199.98	0.1220	0.1221

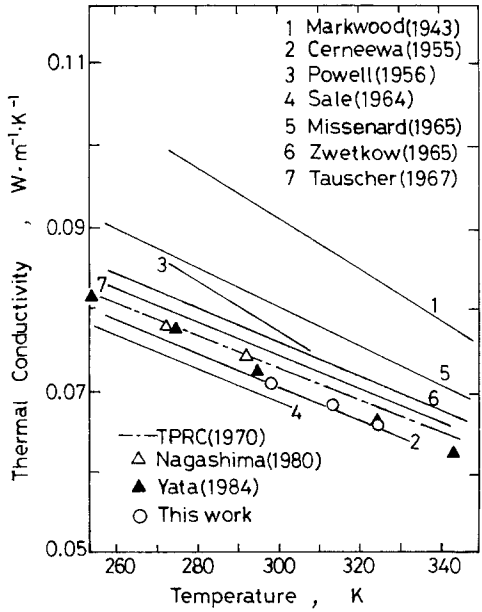

Fig. 1. Temperature dependence of the thermal conductivity of R113 at atmospheric pressure. (1) Ref. 3; (2) Ref. 4; (3) Ref. 5; (4) Ref. 6; (5) Ref. 7; (6) Ref. 8; (7) Ref. 9; (—·—) Ref. 10; (△) Ref. 11; (▲) Ref. 12.

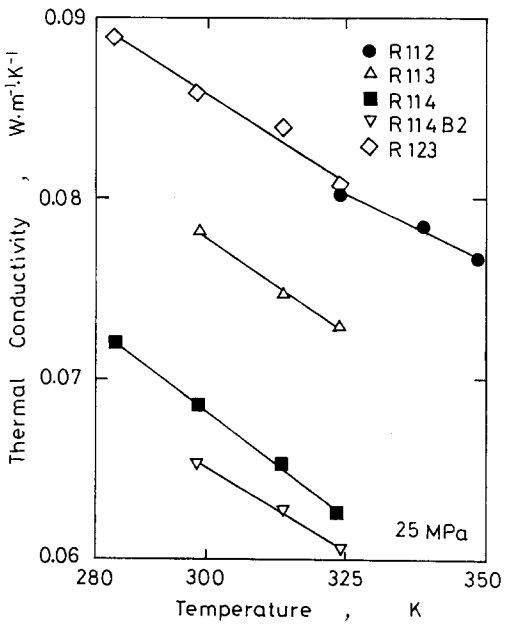

Fig. 2. Isobaric temperature dependence of the thermal conductivity of fluorocarbons at 25 MPa.

In general, the variation of the thermal conductivity of these fluorocarbons with temperature is relatively simple over the entire range in the present work. The thermal conductivity at a constant pressure decreases almost linearly with increasing temperature, that is, the coefficient $(\partial\lambda/\partial T)_P$ is always negative, and its absolute value decreases with increasing pressure.

3.2. Pressure Dependence of Thermal Conductivity

At present, experimental data are quite scarce in the literature for these fluorocarbon liquids under high pressures. In Fig. 3, the isothermal pressure dependence of the thermal conductivity of R113 is typically illustrated as well as the results of Kitazawa and Nagashima [11] by the transient hot-wire method. The maximum deviation of them from ours amounts to nearly 5%.

The pressure dependences of the thermal conductivity of fluorocarbons measured at 313 K are compared in Fig. 4. The pressure coefficient of the thermal conductivity, $(\partial\lambda/\partial P)_T$, increases in the order of R114B2 < R113 < R114 < R112 < R123.

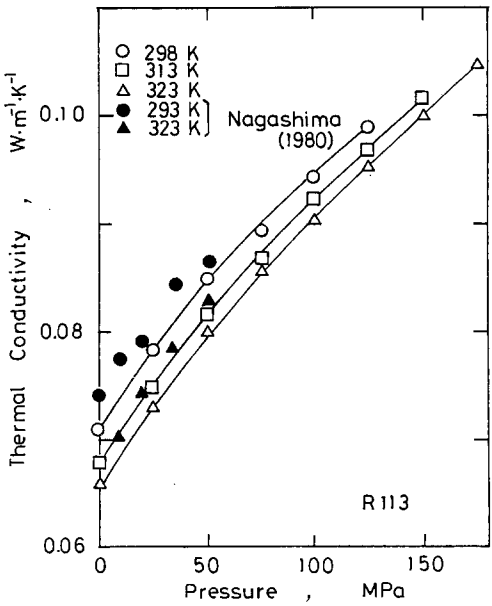

Fig. 3. Isothermal pressure dependence of the thermal conductivity of R113.

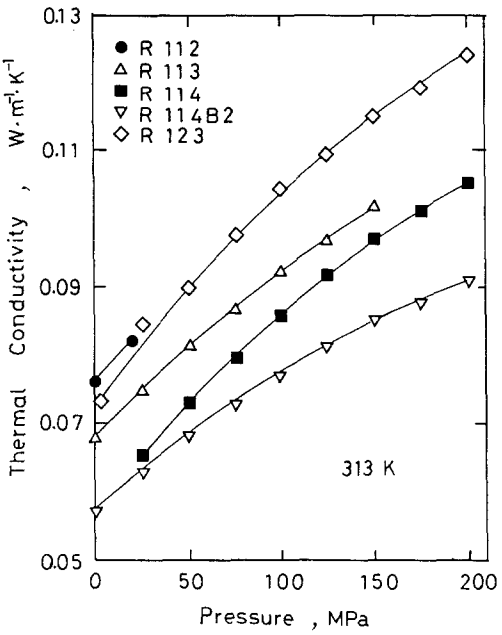

Fig. 4. Isothermal pressure dependence of the thermal conductivity of fluorocarbons at 313 K.

3.3. Density Dependence of Thermal Conductivity

The density dependence of the thermal conductivity of R113, R114, R114B2, and R123 is shown in Fig. 5. The density was measured by the high-pressure burrette method [13] and a vibrating-tube densitometer. The thermal conductivity increases with increasing density. The density coefficient, $(\partial\lambda/\partial\rho)_T$, is always positive and becomes larger with increasing density. The isotherms of each refrigerant seem to depict a single curve independent of temperature. Strictly speaking, however, the density coefficient of λ varies slightly but systematically with temperature.

4. DISCUSSION

4.1. Corresponding-States Correlation

Many thermal conductivity correlations of fluids under high pressures have been based on the corresponding-states principle, wherein either λ/λ^0 or λ/λ_c is expressed as a function of reduced pressure and reduced temperature, where λ^0 is the low-pressure value of λ at the same temperature

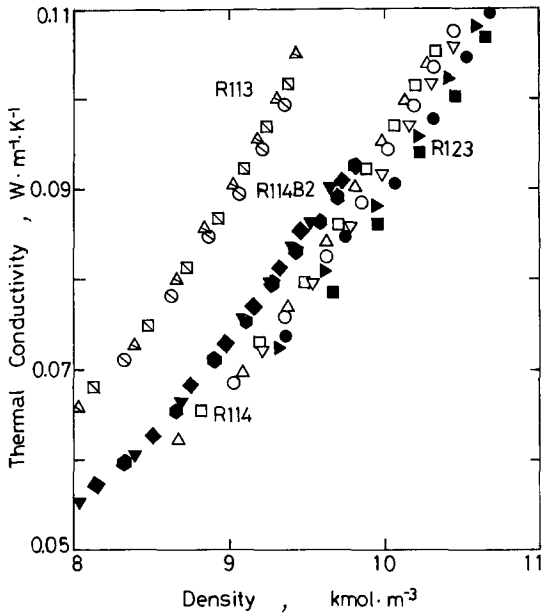

Fig. 5. Isothermal density dependence of the thermal conductivity of R113, R114, R114B2, and R123. R113: (⊘) 298 K; (⊠) 313 K; (△) 323 K. R114B2: (●) 298 K; (◆) 313 K; (▼) 323 K. R114: (▽) 283 K; (○) 293 K; (□) 313 K; (△) 323 K. R123: (■) 298 K; (●) 313 K; (▶) 323 K.

and λ_c is the value at the critical point. In the case of liquids, however, the correlation has been often performed by means of λ itself instead of the reduced thermal conductivity. The present data on the thermal conductivity of R112, R113, and R114 are plotted as a function of reduced temperature and reduced pressure in Fig. 6, where the curves are drawn by interpolation using Eq. (5), described later. Figure 6 suggests that the corresponding-states correlation holds well for these fluorocarbons, although the correlation is not applicable to R114B2 and R123. The present correlation curves can be expressed satisfactorily by the following equations:

$$\lambda = A + BT_r \tag{2}$$

$$A = A_1 + A_2 P_r \tag{3}$$

$$B = B_1 + B_2 P_r + B_3 P_r^2 + B_4 P_r^3 \tag{4}$$

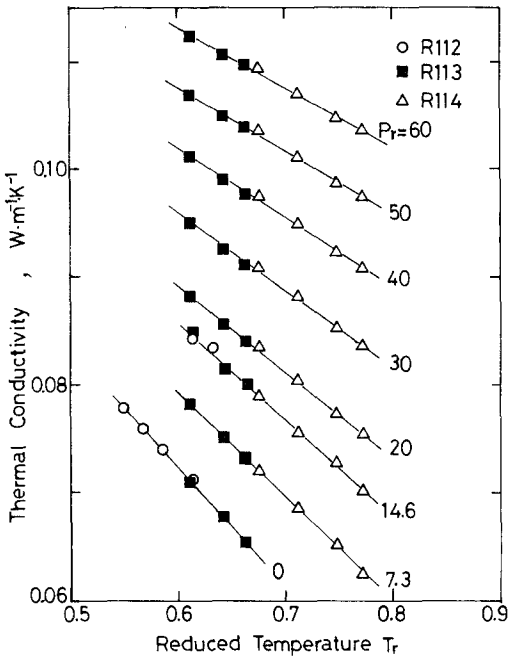

Fig. 6. The corresponding-states correlation of the thermal conductivity of R112, R113, and R114.

where $T_r = T/T_c$ and $P_r = P/P_c$. The critical constants used in this work are as follows:

$$\text{R112: } T_c = 551.0 \text{ K}, \quad P_c = 3.44 \text{ MPa},$$

$$\text{R113: } T_c = 487.2 \text{ K}, \quad P_c = 3.41 \text{ MPa},$$

$$\text{R114: } T_c = 418.78 \text{ K}, \quad P_c = 3.248 \text{ MPa}.$$

The optimum coefficients of Eqs. (3) and (4) are listed in Table VI. The above equations enable us to calculate the thermal conductivity of R112, R113, and R114 with a mean deviation of 1.8% and the maximum of 4.8%. Large deviations are found to occur mainly below 25 MPa.

Latini and Baroncini [14] have proposed a different type of the correlation formulas for the estimation of thermal conductivity based on the corresponding-states principle. Although our data were attempted to correlate by means of their formulas, it is found that systematic deviations occur with increasing pressure.

Table VI. Coefficients in Eqs. (3) and (4)

A_1	1.3578×10^{-1}
A_2	1.3852×10^{-4}
B_1	-1.1027×10^{-1}
B_2	2.1868×10^{-3}
B_3	-3.4427×10^{-5}
B_4	2.2194×10^{-7}
Applicable range of T_r and P_r	$0.55 \leqslant T_r \leqslant 0.77$ $0 \leqslant P_r \leqslant 60$

4.2. The Tait-Type Equation

As described previously [1, 15, 16] the isothermal variation of the liquid thermal conductivity is represented well by an expression similar to the Tait equation as follows:

$$(\lambda - \lambda_0)/\lambda = D \ln[(E+P)/(E+P_0)] \tag{5}$$

where λ_0 is the thermal conductivity at a reference pressure P_0, and P is the pressure in MPa. The present experimental data have been correlated with pressure and temperature by Eq. (5). P_0 is assigned to 0.1 MPa for R113, R114B2, and R123 and 25 MPa for R114, respectively. The empirical coefficients D and E were determined by the nonlinear regression method. Although the optimum value could be determined for each isotherm, it is more convenient to treat D as a characteristic constant of each fluorocarbon. Therefore D was fixed at the mean value, and E was redetermined. The empirical coefficients thus obtained are listed in Table VII together with the deviations of experimental data from Eq. (5). The coefficient E and the thermal conductivity λ_0 are correlated with temperature by the following equations:

$$E = E_1 + E_2 T + E_3 T^2 \tag{6}$$

$$\lambda_0 = a + bT \tag{7}$$

The empirical coefficients are given in Table VIII. Equations (5) to (7) enable us to calculate the thermal conductivity of fluorocarbons studied within the uncertainty of this work, although this correlation is not applied to R112 because it freezes at quite low pressures.

Table VII. Coefficients in Eq. (5)

Temp. (K)	D	E (MPa)	λ_0 ($W \cdot m^{-1} \cdot K^{-1}$)	Mean dev. (%)	Max. dev. (%)
			R113: $P_0 = 0.1$ MPa		
298.15	0.225	49.33	0.0706	0.27	0.77
313.15	0.225	44.56	0.0678	0.26	0.64
323.15	0.225	40.58	0.0653	0.22	0.43
			R114: $P_0 = 25.0$ MPa		
283.15	0.226	23.41	0.0721	0.25	0.60
298.15	0.226	18.74	0.0686	0.32	0.58
313.15	0.226	14.28	0.0653	0.42	0.71
323.15	0.226	10.94	0.0626	0.67	1.34
			R114B2: $P_0 = 0.1$ MPa		
298.15	0.211	44.75	0.0595	0.42	0.54
313.15	0.211	40.41	0.0569	0.43	0.96
323.15	0.211	36.58	0.0546	0.42	1.34
			R123: $P_0 = 0.1$ MPa		
283.15	0.190	30.92	0.0791	0.22	0.43
298.15	0.190	27.08	0.0752	0.25	0.77
313.15	0.190	25.02	0.0722	0.52	1.47
323.15	0.190	20.49	0.0674	0.54	1.81

4.3. Correlation by Dense Hard-Sphere Theory

Based on the hard-sphere theory developed by Dymond [17, 18], Palavra et al. [19] and Li et al [20] employed the following reduced thermal conductivity:

$$\begin{aligned} \lambda' &= (\lambda/\lambda^0)(V/V_0)^{2/3} \\ &= 1.936 \times 10^7 \lambda V^{2/3}(M/RT)^{1/2} \end{aligned} \tag{8}$$

Table VIII. Coefficients of Eqs. (6) and (7)

Substance	E_1 (MPa)	E_2 ($MPa \cdot T^{-1}$)	$10^3 E_3$ ($MPa \cdot T^{-2}$)	a ($W \cdot m^{-1} \cdot K^{-1}$)	$10^3 b$ ($W \cdot m^{-1} \cdot K^{-1} \cdot T^{-1}$)
R113	153.0	−0.3474		0.1378	−0.2240
R114	83.08	−0.1249	−0.3039	0.1382	−0.2333
R114B2	141.5	−0.3240		0.1172	−0.1933

Table IX. The Ratio of $V_0(T)/V_0(298.15)$ in Eq. (8)

Temp. (K)	R113	R114	R114B2
283.15		1.0016	
298.15	1.0000	1.0000	1.0000
313.15	0.9931	0.9987	0.9968
323.15	0.9925	0.9955	0.9848

where λ^0 is the thermal conductivity of dilute gas, V_0 the molar volume of close packing for hard spheres, M the molecular weight, and R the gas constant. This expression has been examined for the present results.

When the quantity λ' is plotted against $\ln V$, the isotherm is almost linear, and its slope is similar at each temperature. Therefore the isotherms can be superimposed into a single curve merely by imposing relative shift along the $\ln V$ axis only. In order to construct this superposition, the volume V_0 at 298.15 K was fixed arbitrarily to 1.0000 for each substance. Shifts of the lines λ' vs $\ln V$ at other temperatures were applied to achieve coincidence. Subsequently, the optimum value of the ratio $V_0(T)/V_0$ (298.15 K) was determined at each temperature for each refrigerant, as given in Table IX.

The quantity $\ln \lambda'$ is found to be expressed by a linear equation of $\ln(V/V_0)$ independent of temperature as follows:

$$\ln \lambda' = c + d \ln (V/V_0) \tag{9}$$

The coefficients c and d are listed in Table X as well as the deviations of the experimental data from Eq. (8).

5. CONCLUSION

As a continuation of the study of the thermophysical properties of fluorocarbons in our laboratory, we have measured the thermal conduc-

Table X. Coefficients in Eq. (9)

Substance	c	d	Mean dev. (%)	Max. dev. (%)
R113	−16.62	−2.214	0.19	0.49
R114	−19.44	−2.488	0.22	0.61
R114B2	−14.92	−2.024	0.20	0.62

tivity of liquid halogenated ethanes R112, R113, R114, R114B2, and R123 at temperatures from 283 to 348 K under pressures up to 200 MPa or freezing pressures by a transient hot-wire method. The thermal conductivity obtained has been correlated satisfactorily with pressure, temperature, and molar volume by the corresponding-states principle, Eq. (2), and other empirical methods, Eqs. (5) and (8). The experimental results of R123 would contribute to the selection of alternatives to the chlorofluorocarbons.

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