

The Thermal Conductivity of R22, R142b, R152a, and Their Mixtures in the Liquid State¹

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An experimental apparatus for measuring the thermal conductivity of liquids by the transient hot-wire method was constructed and tested with toluene as a standard liquid. Measurements were performed on R22, R142b, and R152a. The thermal conductivities of mixtures of R142b and R152a with R22 were also measured by varying the weight fraction of R22. Experiments were performed in the range from -50 to 50°C and from 2 to 20 MPa and the measured data are analyzed to obtain a correlation in terms of temperature, pressure, and composition of the mixture. While the thermal conductivity of R22 + R152a mixtures varies monotonously with composition, that of R22 + R142b mixtures turned out to go through an extremum value. The accuracy of our measurements is estimated to be within 2%.

KEY WORDS: mixtures; R142b; R152a; R22; refrigerants; thermal conductivity; transient hot-wire method.

1. INTRODUCTION

Many thermodynamic systems such as heat pumps and refrigerators operate with CFCs as working fluids. An optimum design of systems involving heat transfer requires a good knowledge of transport properties (thermal conductivity and viscosity) of these working fluids. Many investigations have been carried out to tabulate these properties for various CFCs. However, because of the depletion of the ozone layer in the atmosphere, commercial use of many popular CFCs is now subject to regulation in many countries. As a result, it has become necessary to develop alternative refrigerants and to use mixtures of unregulated refrigerants, the

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performances of which should be comparable to those of the conventional CFCs. Reliable thermal-conductivity data for such refrigerants and their mixtures are not sufficiently available.

It has been known that the measurement of thermal conductivity of refrigerants is a field in which many of the reported values are considerably scattered [1–5]. Such a discord among experimental data is due mainly to the fact that refrigerants, unlike standard fluids such as toluene and *n*-heptane, are electrically conductive.

Recently, many researchers have tried to obtain accurate thermal-conductivity data for refrigerants. Thermal-conductivity data at the saturated liquid state have been reported by Yata et al. [2] for 10 liquid fluorocarbons and by Fellows et al. [3] for several environmentally acceptable fluorocarbons. Assael et al. [4, 5] reported thermal conductivities of several refrigerants in a wide range of temperature and pressure. They used anodized tantalum wires to reduce the effect of electrical conductivity of fluorocarbons. An examination of available data compilation shows that thermal conductivities of many alternative refrigerants, especially of R141b, R142b, and R152a, are scarce and that few experimental data for refrigerant mixtures exist.

The main objective of this study is to provide thermal-conductivity data for a few refrigerants (R22, R142b, and R152a) and for refrigerant mixtures (R22 + R142b and R22 + R152a). We employ the transient hot-wire method that is known to reduce the effect of convective heat transfer. The measurement uncertainty in our experiments is estimated to be $\pm 2\%$. For all the refrigerants considered, measurements have been performed in a range of temperature from -50 to 50°C and in a range of pressure from 2 to 20 MPa. In the case of the mixtures, the weight fraction of R22 plays a role as an additional independent parameter so that the total number of data exceeds 200. For an assessment of our measured data, subsidiary experiments have been conducted with toluene at temperatures ranging from 20 to 60°C , and when compared with the data reported by Nieto de Castro et al. [6], agreement to within 1% is achieved.

2. BASIC CONSIDERATIONS

The transient hot-wire method is one of the unsteady state techniques that yields the thermal conductivity via the measurement of a timewise temperature variation of the hot-wire. An in-depth description of this technique is given, among others, by de Groot et al. [7].

The basic theory to determine the thermal conductivity begins with the following equations:

$$\Delta T(a, t) = \frac{q}{4\pi\lambda(T_r, P)} \ln\left(\frac{4\alpha t}{a^2 C}\right) \quad (1)$$

$$T_r = T_0 + \frac{1}{2} \{ \Delta T(a, t_1) + \Delta T(a, t_2) \} \quad (2)$$

where $\Delta T(a, t)$ denotes the surface temperature rise of the wire of radius a at a certain time t , q the heat flux supplied per unit length of the wire, $\lambda(T_r, P)$ the thermal conductivity of interest, P the pressure of the medium, and T_0 the initial equilibrium temperature at $t=0$, t_1 and t_2 designate two instant times after the onset of the experiment, and C is the exponential of Euler's constant [$C = \exp(0.5772\dots)$]. With a known amount of heat supply q , the thermal conductivity is obtained from Eq. (1) by measuring the temperature change ΔT during the time interval. The initial and final times of measurement are selected to minimize the discrepancy between the theory and the actual measurement.

In our experiments, only one wire is installed in the hot-wire cell and no attempt has been made to compensate the end-effect error. As for the conductive and convective end effects, Knibbe [8] set forth an error criterion that was provisionally labeled as "the worst-case analysis." Based on his analysis, the maximum end-effect error in our measurement is found to be less than 0.5% for the case of toluene, which appears acceptable for our measuring purpose.

When a bare platinum wire is used for electrically conducting refrigerants, there will occur a certain amount of current leakage and polarization, which can be directly detected from the distortion of the linearity between ΔT and the logarithm of time. Another effect of current leakage is the alteration of the constant current through the wire. In order to quantify the influence of current leakage and polarization, contaminated water with a higher electric conductivity than refrigerants was tested and compared against the case with pure water (data from Nieto de Castro et al. [6]). It was found that the occurrence of current leakage lowered the thermal conductivity about 4–5%. A question whether such a deviation increases linearly with the magnitude of current leakage still remains unsolved. However, it is expected that the cases with refrigerants are less affected by current leakage than the case with the contaminated water. In this respect, Fellows et al. [3] measured thermal conductivities of liquid refrigerants and presumed the accuracy of their experiments to be 2–3% with current leakage taken into consideration. It may be postulated that our estimation agrees to theirs to a similar order.

The temperature gradient imposed by heating the wire leads to variation of composition (i.e., thermal-diffusive separation of components) in

mixtures. Khalifa et al. [9] studied this topic for the case of gas mixtures. They showed that no attention needs to be paid to composition variation when using a transient hot-wire method. Also, Fareleira et al. [10] have asserted that the same reasoning holds for the case of liquid mixtures. Therefore, the effect of thermal-diffusive separation is not considered in this study.

3. EXPERIMENTAL APPARATUS

A schematic diagram describing the pressure vessel and the hot-wire cell is shown in Fig. 1. The pressure vessel is composed of a main body and a cover all of which are made of SUS 304. The cover has an outer diameter of 180 mm and a thickness of 23 mm. Near the center of the cover, three small holes are punched out; two for current supply and one for measuring voltage signal. A Pt 100- Ω -resistance thermometer is embedded deep into the pressure vessel and fixed to the cover by welding. The main body has

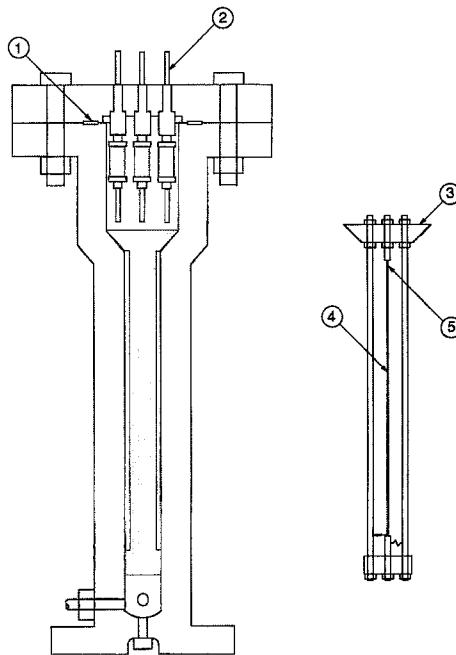


Fig. 1. Schematic diagram of the pressure vessel and the hot-wire cell. 1, Copper gasket; 2, electric conductor; 3, Teflon stator; 4, platinum wire; 5, spring.

a height of 315 mm and an inner diameter of 28 mm. The diameter of the platinum wire is 25 μm . The initial resistance of the hot wire is obtained by supplying 1 mA and then by measuring the voltage via the four-wire method with the precise potentiometer.

Each sample fluid is pressurized with a liquid pump. Internal pressure is then measured with a bourdon-type pressure gauge with an accuracy of 0.4% (full-scale range) and no pressure change is found during a 24-h leak test. To maintain the sample fluid at a constant temperature, a constant temperature bath which encloses the pressure vessel is used and its temperature is controlled with a 1-kW heater and a temperature controller. To check the thermal equilibrium between the bath and the vessel, the bath temperature is measured and compared with the vessel temperature. The temperature fluctuation of the bath is found to be less than 0.1°C.

The operation of both the temperature controller and the refrigerator gives rise to the electric noise during the measurement, and the mechanical vibration is also responsible for the early onset of natural convection. Therefore, during the data acquisition, the temperature-control system is turned off.

The electric system is composed of a Wheatstone-bridge circuit and a data-processing system. A constant current is supplied to the Wheatstone bridge by a constant-current supplier. The out-of-balance-signal of the bridge is amplified by a differential amplifier and converted to a digital signal by an A/D converter which has a 12-bit accuracy. The differential amplifier and the A/D converter are calibrated as a whole by measuring the amplification ratio, stability and the time constant with a constant current supplied to standard resistor. The sampling frequency of our measurement is 2 kHz and the temperature rise during the measurement is about 1.5°C.

4. THERMAL CONDUCTIVITY OF TOLUENE

For the verification of our measurement, the thermal conductivity of toluene of commercial grade purity (above 98%) is measured in a tem-

Table I. Thermal Conductivity of Toluene at Atmospheric Pressure

Temperature (°C)	Thermal conductivity ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)
20.0	0.1315
40.0	0.1260
60.0	0.1204

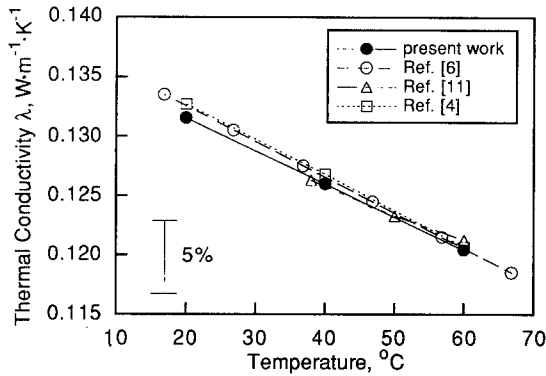


Fig. 2. Thermal conductivity of toluene at atmospheric pressure.

perature range of 20–60°C at atmospheric pressure. The results are listed in Table I. Figure 2 compares our data with those of other researchers. It can be seen that the present data are in good agreement (to within 1%) with those of others.

5. RESULTS

The thermal conductivity has been measured for R22, R142b, R152a, and their mixtures in the range of -50 – 50 °C and 2–20 MPa. The purities of R22, R142b, and R152a are better than 99.5, 99.8, and 99.8%, respectively, by manufacturers' analysis. For each combination of temperature and pressure, measurements are repeated five times and the reproducibility is observed to be within $\pm 0.5\%$. The measured values are then corrected to the nominal temperatures at the experimental pressure in order to examine the composition dependence of the thermal conductivity. The average value of these corrections is 0.36% with a maximum value of 1.44%. Table II lists the thermal-conductivity data of R22 + R142b mixtures for several weight fractions of R22. Also listed in Table III are the thermal-conductivity data of R22 + R152a mixtures for various weight fractions of R22. The experimental data for the thermal conductivity of pure refrigerants have been fitted to an equation of the form:

$$\lambda = \lambda_0 \sum_{j=0}^2 \sum_{i=0}^2 a_{ij} T^i P^j \quad (3)$$

with T in °C and P in MPa. The values for the coefficients λ_0 and a_{ij} are listed in Table IV for R22, R142b and R152a. The average deviations of

the experimental thermal-conductivity data for R22, R142b, and R152a from Eq. (3) are 0.22, 0.29, and 0.11%, respectively, and the maximum deviations do not exceed 0.52, 0.85, and 0.28%, respectively.

Figure 3 shows the pressure dependence of the thermal conductivity of pure refrigerants along isotherms, and Fig. 4 the temperature dependence along isobars. As evidenced in Figs. 3 and 4, the pressure dependence of the thermal conductivity deviates to some extent from a linear relation at low pressure and at high temperature.

To compare our data with those of other researchers, the measured thermal conductivities have been extrapolated from Eq. (3) to the saturation pressure of each refrigerant at a given temperature. Figure 5 displays the temperature dependence of thermal conductivity of saturated liquid

Table II. Measured Thermal Conductivity of R22 + R142b Mixtures

T_{nom} (°C)	P (MPa)	Thermal conductivity λ (W · m ⁻¹ · K ⁻¹) at a weight fraction of R22 of				
		0.0000	0.2796	0.4350	0.7290	1.0000
-50.0	2.1	0.1124	0.1091	0.1107	0.1141	0.1189
-50.0	5.1	0.1134	0.1101	0.1118	0.1151	0.1201
-50.0	10.1	0.1150	0.1120	0.1137	0.1171	0.1223
-50.0	15.1	0.1167	0.1139	0.1155	0.1190	0.1244
-50.0	20.1	0.1183	0.1154	0.1174	0.1207	0.1262
-25.0	2.1	0.1013	0.0990	0.0997	0.1030	0.1069
-25.0	5.1	0.1033	0.1003	0.1010	0.1042	0.1086
-25.0	10.1	0.1053	0.1023	0.1033	0.1064	0.1112
-25.0	15.1	0.1077	0.1046	0.1057	0.1086	0.1138
-25.0	20.1	0.1098	0.1065	0.1077	0.1106	0.1157
0.0	2.1	0.0914	0.0892	0.0898	0.0915	0.0956
0.0	5.1	0.0933	0.0908	0.0915	0.0934	0.0974
0.0	10.1	0.0958	0.0935	0.0940	0.0961	0.1006
0.0	15.1	0.0976	0.0955	0.0964	0.0987	0.1032
0.0	20.1	0.1001	0.0978	0.0985	0.1010	0.1063
25.0	2.1	0.0817	0.0801	0.0800	0.0813	0.0842
25.0	5.1	0.0833	0.0820	0.0820	0.0831	0.0866
25.0	10.1	0.0857	0.0845	0.0847	0.0866	0.0903
25.0	15.1	0.0891	0.0872	0.0875	0.0895	0.0936
25.0	20.1	0.0909	0.0897	0.0900	0.0921	0.0967
50.0	2.1	0.0734	0.0712	0.0706	0.0707	0.0713
50.0	5.1	0.0750	0.0730	0.0729	0.0732	0.0748
50.0	10.1	0.0781	0.0764	0.0764	0.0772	0.0791
50.0	15.1	0.0809	0.0794	0.0796	0.0808	0.0831
50.0	20.1	0.0840	0.0822	0.0819	0.0837	0.0869

Table III. Measured Thermal Conductivity of R22 + R152a Mixtures

T_{nom} (°C)	P (MPa)	Thermal conductivity λ (W · m ⁻¹ · K ⁻¹) at a weight fraction of R22 of				
		0.0000	0.2488	0.4968	0.7505	1.000
-50.0	2.1	0.1376	0.1324	0.1274	0.1233	0.1189
-50.0	5.1	0.1391	0.1334	0.1284	0.1243	0.1201
-50.0	10.1	0.1412	0.1353	0.1308	0.1264	0.1223
-50.0	15.1	0.1432	0.1376	0.1329	0.1284	0.1244
-50.0	20.1	0.1450	0.1391	0.1350	0.1303	0.1262
-25.0	2.1	0.1252	0.1193	0.1153	0.1108	0.1069
-25.0	5.1	0.1267	0.1215	0.1170	0.1122	0.1086
-25.0	10.1	0.1296	0.1240	0.1196	0.1148	0.1112
-25.0	15.1	0.1321	0.1266	0.1218	0.1175	0.1138
-25.0	20.1	0.1344	0.1282	0.1240	0.1195	0.1157
0.0	2.1	0.1140	0.1086	0.1040	0.0990	0.0956
0.0	5.1	0.1155	0.1107	0.1054	0.1005	0.0974
0.0	10.1	0.1181	0.1133	0.1082	0.1035	0.1006
0.0	15.1	0.1212	0.1161	0.1114	0.1065	0.1032
0.0	20.1	0.1241	0.1188	0.1139	0.1089	0.1063
25.0	2.1	0.1029	0.0968	0.0919	0.0870	0.0842
25.0	5.1	0.1049	0.0991	0.0944	0.0898	0.0866
25.0	10.1	0.1081	0.1026	0.0976	0.0931	0.0903
25.0	15.1	0.1116	0.1057	0.1006	0.0963	0.0936
25.0	20.1	0.1142	0.1089	0.1040	0.0992	0.0967
50.0	2.1	0.0928	0.0856	0.0808	0.0759	0.0713
50.0	5.1	0.0954	0.0884	0.0832	0.0788	0.0748
50.0	10.1	0.0986	0.0928	0.0873	0.0832	0.0791
50.0	15.1	0.1018	0.0967	0.0911	0.0871	0.0831
50.0	20.1	0.1052	0.0994	0.0946	0.0900	0.0869

Table IV. Coefficients in Eq. (3) for R22, R142b, and R152a

	R22	R142b	R152a
λ_0	9.4261×10^{-2}	9.0103×10^{-2}	1.1250×10^{-1}
a_{00}	1.0000×10^0	1.0000×10^0	1.0000×10^0
a_{01}	6.9981×10^{-3}	6.5290×10^{-3}	5.2488×10^{-3}
a_{02}	-3.4812×10^{-5}	-5.0025×10^{-5}	-5.3822×10^{-6}
a_{10}	-5.0957×10^{-3}	-4.4070×10^{-3}	-4.0113×10^{-3}
a_{11}	5.9622×10^{-5}	2.4517×10^{-5}	2.1447×10^{-5}
a_{12}	-6.3793×10^{-7}	6.3354×10^{-8}	7.9389×10^{-8}
a_{20}	-3.1958×10^{-6}	8.0651×10^{-6}	5.2782×10^{-6}
a_{21}	5.7502×10^{-7}	-6.0837×10^{-7}	1.5425×10^{-7}
a_{22}	-1.7076×10^{-8}	2.2108×10^{-8}	-1.2686×10^{-8}

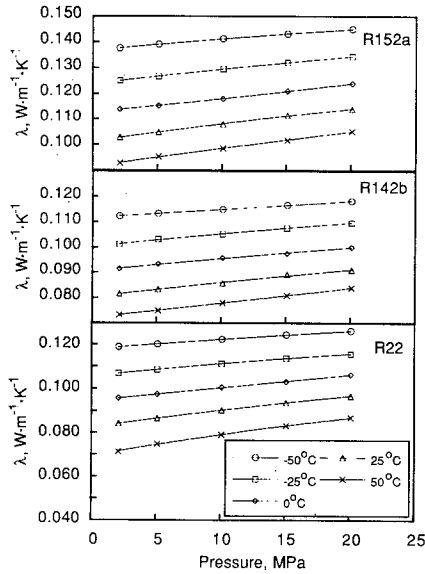


Fig. 3. Measured thermal conductivities of R22, R142b, and R152a; pressure dependence along isotherms.

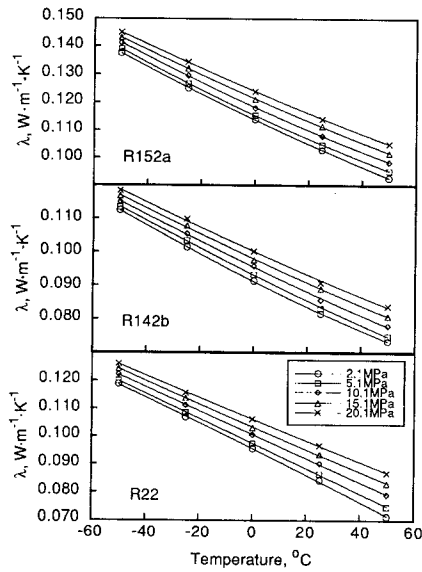


Fig. 4. Measured thermal conductivities of R22, R142b, and R152a; temperature dependence along isobars.

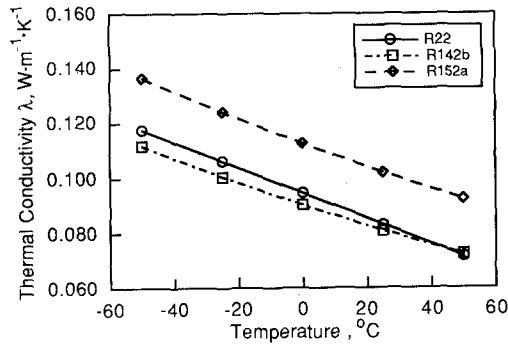


Fig. 5. Extrapolated thermal conductivities of R22, R142b, and R152a at the saturated liquid state.

Table V. Thermal Conductivity of R22, R142b, and R152a at the Saturated Liquid State

Temperature (°C)	Pressure (MPa)	Thermal conductivity (W · m ⁻¹ · K ⁻¹)	Difference (%) ^a				
			δ^b	δ^c	δ^d	δ^e	δ^f
R22							
-50.0	0.0645	0.1176					
-25.0	0.2016	0.1062	1.7				
0.0	0.4979	0.0946	1.5		-0.1		
25.0	1.0444	0.0829	1.4		-0.4		
50.0	1.9432	0.0715	0.8	4.0	-0.8		
R142b							
-50.0	0.0156	0.1118					1.6
-25.0	0.0536	0.1005				2.6	4.1
0.0	0.1466	0.0902				3.8	5.3
25.0	0.3387	0.0808				5.3	4.8
50.0	0.6871	0.0725				6.8	2.0
R152a							
-50.0	0.0309	0.1366					4.5
-25.0	0.1013	0.1242				2.9	3.9
0.0	0.2669	0.1127				3.0	2.4
25.0	0.5981	0.1020				3.3	-0.3
50.0	1.1822	0.0923				3.8	-4.7

^a Difference, δ , is calculated from Eq. (4).

^b Ref. 2.

^c Ref. 3.

^d Ref. 5.

^e Johns (1983) in Ref. 12.

^f Ref. 13.

Table VI. Coefficients in Eq. (5) for R22 + R142b Mixtures

λ_0	8.9990×10^{-2}		
	$k=0$	$k=1$	$k=2$
a_{00k}	1.0000×10^0	-1.0520×10^{-1}	1.5359×10^{-1}
a_{01k}	6.4878×10^{-3}	-2.4275×10^{-3}	3.3072×10^{-3}
a_{02k}	-4.7605×10^{-5}	6.1654×10^{-5}	-5.3657×10^{-5}
a_{10k}	-4.3511×10^{-3}	1.5753×10^{-4}	-1.1884×10^{-3}
a_{11k}	2.0641×10^{-5}	7.8280×10^{-5}	-3.4629×10^{-5}
a_{12k}	2.0928×10^{-7}	-3.4919×10^{-6}	2.5793×10^{-6}
a_{20k}	7.3067×10^{-6}	-8.5368×10^{-6}	-1.4733×10^{-6}
a_{21k}	-5.5458×10^{-7}	2.3353×10^{-6}	-1.2397×10^{-6}
a_{22k}	2.0542×10^{-8}	-8.2053×10^{-8}	4.5952×10^{-8}

refrigerants. Table V lists the differences between the present data and other reference data. The difference δ appearing in Table V is calculated as

$$\delta = \frac{\lambda_{\text{ref.}} - \lambda_{\text{meas.}}}{\lambda_{\text{meas.}}} \times 100(\%) \tag{4}$$

Figure 5 reveals that, over the entire range of temperature examined, R152a has the highest thermal conductivity and that the temperature dependence is the most severe for the case of R22.

Table VII. Coefficients in Eq. (5) for R22 + R152a Mixtures

λ_0	1.1251×10^{-1}		
	$k=0$	$k=1$	$k=2$
a_{00k}	1.0000×10^0	-2.0870×10^{-1}	4.5527×10^{-2}
a_{01k}	5.3896×10^{-3}	8.9015×10^{-4}	-4.5071×10^{-4}
a_{02k}	-1.1512×10^{-5}	-6.7965×10^{-5}	5.1585×10^{-5}
a_{10k}	-4.0266×10^{-3}	-7.2004×10^{-4}	4.7360×10^{-4}
a_{11k}	2.4129×10^{-5}	7.1230×10^{-5}	-4.4017×10^{-5}
a_{12k}	4.1165×10^{-8}	-2.3171×10^{-6}	1.6396×10^{-6}
a_{20k}	4.2883×10^{-6}	-6.2450×10^{-6}	3.3686×10^{-7}
a_{21k}	2.1857×10^{-7}	-1.1822×10^{-7}	3.3994×10^{-7}
a_{22k}	-1.4639×10^{-8}	3.3710×10^{-8}	-3.2870×10^{-8}

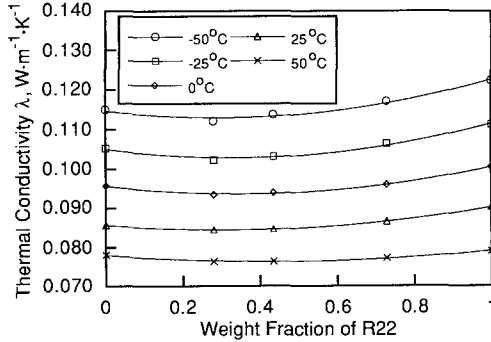


Fig. 6. Measured thermal conductivities of R22 + R142b mixtures; composition dependence along isotherms for $P = 10.1 \text{ MPa}$.

The experimental data for the thermal conductivity of refrigerant mixtures have been fitted to an equation of the form

$$\lambda = \lambda_0 \sum_{k=0}^2 \sum_{j=0}^2 \sum_{i=0}^2 a_{ijk} T^i P^j w^k \tag{5}$$

with T in $^{\circ}\text{C}$, P in MPa, while w denotes the weight fraction of R22. The values for the coefficients λ_0 and a_{ijk} are listed in Tables VI and VII for R22 + R142b and R22 + R152a mixtures, respectively. The average deviations of experimental thermal-conductivity data for R22 + R142b and R22 + R152a mixtures from Eq. (5) are 0.26 and 0.12%, respectively and the maximum deviations do not exceed 0.83 and 0.73%, respectively.

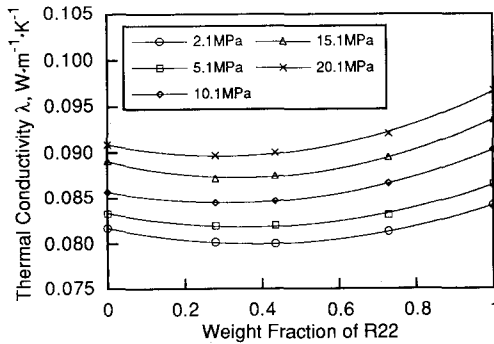


Fig. 7. Measured thermal conductivities of R22 + R142b mixtures; composition dependence along isobars for $T_{\text{nom}} = 25.0^{\circ}\text{C}$.

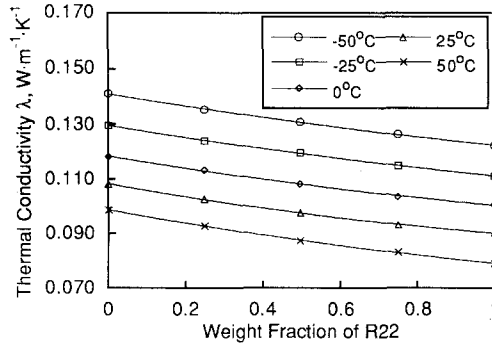


Fig. 8. Measured thermal conductivities of R22 + R152a mixtures; composition dependence along isotherms for $P = 10.1 \text{ MPa}$.

Figure 6 exhibits the isothermal composition dependence of R22 + R142b mixtures for an isobar, and Fig. 7 the isobaric composition dependence for an isotherm. The curved lines represent quadratic fitting of the measured data. In the case of R22 + R142b mixtures, the composition dependence deviates strongly from linearity. The deviations amount to as much as 4.3% at the lowest pressure. For the case of R22 + R152a mixtures, Fig. 8 exposes the isothermal composition dependence of thermal conductivity for an isobar, and Fig. 9 the isobaric composition dependence for an isotherm. When compared to the results shown in Figs. 6 and 7, the composition dependence is not so pronounced. The deviations amount to as much as 2.1% at the lowest pressure. We note that the thermal conductivity of R22 + R142b mixtures falls below that of each pure refrigerant.

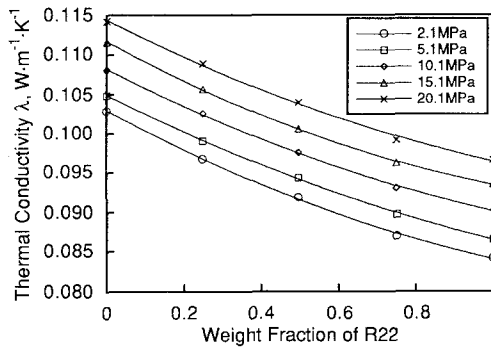


Fig. 9. Measured thermal conductivities of R22 + R152a mixtures; composition dependence along isobars for $T_{\text{nom}} = 25.0^\circ\text{C}$.

It is also very interesting to note that the thermal conductivity of R22 + R142b mixtures has an extremum, whereas that of R22 + R152a mixtures varies monotonously.

6. CONCLUSIONS

An experimental apparatus for measuring the thermal conductivity of liquids by a transient hot-wire method has been constructed and calibrated to measure thermal conductivities of R22, R142b, R152a, and their mixtures in wide ranges of pressure and temperature.

A preliminary investigation has been carried out to obtain the composition dependence of the thermal conductivity of liquid refrigerant mixtures. The measured data reveal that the composition dependence of the thermal conductivity of R22 + R142b and R22 + R152a mixtures is not at all similar. This observation suggests that much care be taken in predicting the thermal conductivity of liquid refrigerant mixtures from that of the pure components.

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