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Measurements of the Thermal Conductivity of Liquid R32, R124, R125, and R141b

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This paper reports measurements of the thermal conductivity of refrigerants R32, R124, R125, and R141b in the liquid phase. The measurements, covering a temperature range from 253 to 334 K and pressure up to 20 MPa, have been performed in a transient hotwire instrument employing two anodized tantalum wires. The uncertainty of the present thermal-conductivity data is estimated to be ±0.5%. The experimental data have been represented by polynomial functions of temperature and pressure for the purposes of interpolation. A comparison with other recent measurements is also included.

KEY WORDS: high pressure; refrigerants; R32; R124; R125; R141b; thermal conductivity; transient hot-wire technique.

1. INTRODUCTION

During the last three years we have undertaken the task of measuring, as accurately as possible, the thermal conductivity and viscosity of 10 refrigerants in the liquid and vapor phase. The refrigerants considered are R11, R12, R22, R32, R123, R124, R125, R141b, R134a, and R152a. Specifically in relation to the thermal-conductivity measurements in the liquid phase, our measurements on R11, R12 [1], R22, R123, R134a [2], and R152a [3] have already been reported. Hence, the present results on R32, R124, R125, and R141b conclude our measurements in the liquid phase.

The main problem in dealing with refrigerants, especially in the liquid phase, is that most of them are polar fluids and act as very good solvents. Although the existence of small ionic impurities does not significantly influence the thermodynamic and transport properties of the liquids, it

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affects the electrical conduction through them. In the case of the transient hot-wire technique, this effect introduces, an experimental error in the measurements of thermal conductivity when bare wires are employed as the electrical heating source. Two techniques have been so far employed in order to reduce this effect. Accoding to the first technique, a fixed DC polarization voltage is applied between the cell walls and the bare wires [4]. Since however, the value of the thermal conductivity depends on the polarization voltage, this technique is slowly being abandoned [5]. The second technique relies on the electrical insulation of the wire by a very thin film. This is easily achieved by employing tantalum wires and anodizing them *in situ*. Upon anodization a very thin, about 100 nm, electrical insulating film of tantalum pentoxide is formed on the wire surface. The technique, employed successfully for many years, is attributed to the ingenious idea developed by Alloush et al. in 1982 [6]. This technique has been employed in our laboratory for all measurements of the thermal conductivity of polar liquids.

2. EXPERIMENTS

The theory of the transient hot-wire technique and the description of the present experimental installation are given in detail elsewhere [1]. To avoid the introduction of small iron particles, which are usually present in iron sample bottles, a 20-μm stainless-steel filter was employed at the inlet of the pressure vessel. Furthermore, the filling of the pressure vessel was accomplished by liquifying the vapor of each refrigerant into the cell, after long hours of vacuum.

All measurements were performed from just above the saturation pressure, up to 20 MPa along the 253.15, 273.15, 293.15, 313.15, and 333.15 K isotherms (except in the case of R32 and R125, where they were restricted to 313.15 K). The thermal conductivity of toluene and water was measured before and after each liquid, to ensure the continuing good operation of the instrument. These measurements were found to agree with our previously reported measurements within ± 0.1 % [1, 7]. The precision of the instrument is ± 0.1 % while the estimated uncertainty of the measurements, as confirmed by the measurement of the thermal conductivity of toluene, is believed to be ± 0.5 %.

The sample of R32 was supplied by I.C.I. Chemicals and Polymers Ltd., at a nominal purity of 99.98 %. The samples of R124 and R125 were supplied by Du Pont de Nemours International S.A., both at a stated purity of better than 99.95 %, while R141b was supplied by Elf Atochem S.A. at a stated purity of better than 99.9 %. Gas chromatography analysis confirmed these purities.

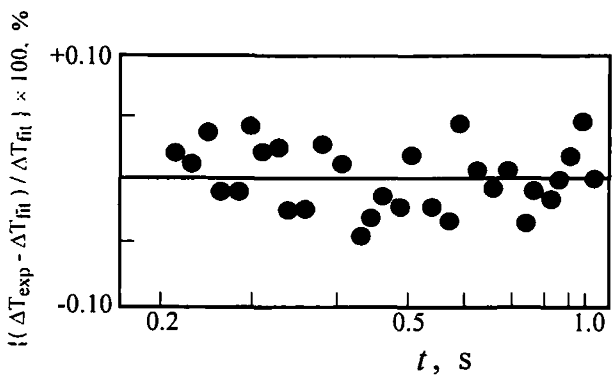

Fig. 1. Percentage deviations of the measured temperature rise as a function of time, from linearity for a typical run of R124 at 333.00 K and 4.75 MPa.

According to the transient hot-wire working equation [8], the measured temperature rise vs the logarithm of the time, should be a straight line. Figure 1 shows the percentage deviations of the experimental temperature rise, ΔT, from the straight line, as a function of time for a typical measurement on R124 at 333.0 K and 4.75 MPa. It can be seen that, not only is there no apparent curvature or systematic trend, but also that the maximum deviation is less than $\pm 0.05\%$. This is taken as conclusive evidence that the instrument behaves according to its working equation.

3. RESULTS

The measurements of the thermal conductivity of the four refrigerants were carried out along five isotherms, 253.15, 273.15, 293.15, 313.15, and 333.15 K (except in the case of R32 and R125, where they were restricted to 313.15 K), from above saturation pressure up to 20 MPa. The density values employed in the calculation of the thermal conductivity of R32 and R125 were calculated from the MBWR equation of state developed by Outcalt and McLinden [9] with a quoted uncertainty of $\pm 0.2\%$. For the density of R124 the only available values, to our knowledge, are the measurements of Kubota et al. [10], with a quoted uncertainty of $\pm 0.09\%$. However, these measurements go down in temperature only to 278 K. Hence for the 253 and 273 K isotherms, we employed the density values given by Diller and Peterson [11], calculated by a corresponding-states model developed by Huber and Ely with an uncertainty of better than $\pm 1\%$.

Finally, in the case of R141b the density values employed were calculated from the equation given by Defibaugh et al. [12], for the temperature range 278–370 K and up to 6.0 MPa, with a quoted uncertainty of $\pm 0.05\%$. Since to our knowledge no other measurements-based equation or measurements of the density of R141b are available, we have extrapolated this equation to lower temperatures and higher pressures. As a further check of this extrapolation, we have compared these values with the corresponding-state density values quoted by Diller et al. [13]. The extrapolated values were found to agree with the latter ones within $\pm 0.7\%$. Sousa and Nieto de Castro [14] have recently presented density measurements for R141b that cover a much wider temperature and pressure range with an uncertainty of $\pm 0.1\%$. These measurements, however, differ from the Defibaugh et al. [12] values by 2%, which is far apart from the mutual uncertainty of the two sets. Moreover, since these measurements have not been published yet, they were not taken into consideration.

It should be pointed out, however, that an error of $\pm 1\%$ in density results in an error of $\pm 0.02\%$ in thermal conductivity. Hence the uncertainty introduced by the extrapolated-density values is negligible.

Tables I to IV show the present experimental measurements of the thermal conductivity of R32, R124, R125, and R141b, respectively. All thermal-conductivity measurements of each refrigerant have been correlated as a function of the reduced temperature, T_r ($=T/T_c$, where T_c is the critical temperature), and reduced pressure, P_r ($=P/P_c$, where P_c is the critical pressure), for the purpose of interpolation only, by an equation of the form

$$\lambda = \sum_{i=0}^{2} \sum_{j=0}^{2} C_{ij} P_r^i T_r^j \tag{1}$$

The values of all constants are shown in Table V. Critical constants shown in the table were obtained from the literature—R32 [15], R124 [10], R125 [16], and R141b [12]. The maximum deviation of all thermal-conductivity measurements from Eq. (1) is 0.15%. In the same table the standard deviation of each fit is also shown. It can be seen that the largest standard deviation is 0.06%. It should also be pointed out that the above equation was employed to calculate the values at nominal temperatures, shown also in Tables I to IV, together with the experimental values.

In Table VI, the thermal conductivity at saturation conditions is shown for the four refrigerants. These values have been obtained by the use of Eq. (1), while values for the saturation pressure, P_s, and saturation density, ρ_s, are obtained from the respective density references discussed previously.

Table I. Measurements of Thermal Conductivity of R32

Pressure P (MPa)	Temperature T (K)	Thermal conductivity $\lambda(T, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$	Density $\rho(T_{nom}, P)$ $(kg \cdot m^{-3})$	Thermal conductivity $\lambda(T_{nom}, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$
		$T_{nom} = 253.15$ K		
0.63	252.60	159.3	1123	158.8
2.78	252.70	160.6	1128	160.2
5.07	252.67	162.2	1134	161.8
8.17	252.69	164.3	1142	163.9
11.05	252.76	166.1	1148	165.8
14.05	252.75	168.2	1155	167.9
17.25	252.90	170.4	1161	170.2
		$T_{nom} = 273.15$ K		
1.10	273.03	142.3	1057	142.2
3.68	272.97	144.8	1067	144.7
6.13	272.90	146.9	1075	146.7
9.18	272.96	149.4	1084	149.3
11.95	272.92	151.6	1093	151.4
14.70	273.00	153.5	1100	153.4
16.93	273.01	154.7	1106	154.6
		$T_{nom} = 293.15$ K		
1.80	292.83	126.2	985	125.9
4.12	292.82	129.1	997	128.8
6.12	292.83	131.5	1007	131.3
8.53	292.80	134.0	1018	133.8
11.18	292.90	136.4	1028	136.2
13.90	293.11	138.5	1037	138.5
17.30	293.15	141.1	1049	141.1
		$T_{nom} = 313.15$ K		
2.84	312.55	111.1	900	110.6
5.12	312.50	114.8	920	114.3
7.71	312.65	118.5	938	118.1
10.00	312.58	121.8	953	121.4
12.88	312.56	125.3	968	124.9
17.63	312.83	129.8	989	129.6

Table II. Measurements of Thermal Conductivity of R124

Pressure P (MPa)	Temperature T (K)	Thermal conductivity $\lambda(T, P)$ ($mW \cdot m^{-1} \cdot K^{-1}$)	Density $\rho(T_{nom}, P)$ ($kg \cdot m^{-3}$)	Thermal conductivity $\lambda(T_{nom}, P)$ ($mW \cdot m^{-1} \cdot K^{-1}$)
		$T_{nom} = 253.15$ K		
0.62	252.94	83.5	1486	83.4
2.35	252.93	84.3	1491	84.2
5.90	252.84	85.9	1502	85.8
8.94	252.75	87.2	1511	87.1
12.11	253.01	88.4	1519	88.4
15.21	252.90	89.7	1528	89.6
18.06	252.40	90.9	1535	90.7
		$T_{nom} = 273.15$ K		
0.64	273.23	76.7	1432	76.7
2.70	273.17	77.8	1440	77.8
6.08	273.12	79.4	1452	79.4
9.18	273.12	80.9	1463	80.9
12.32	273.16	82.3	1474	82.3
15.34	273.16	83.6	1483	83.6
18.67	273.21	85.0	1493	85.0
		$T_{nom} = 293.15$ K		
0.63	293.33	70.1	1378	70.2
3.23	293.07	71.7	1390	71.7
5.43	293.25	72.9	1399	72.9
6.17	293.04	73.3	1402	73.3
9.19	293.10	74.9	1414	74.9
12.18	293.05	76.4	1424	76.4
15.19	293.05	77.9	1435	77.9
18.16	293.09	79.2	1444	79.2
		$T_{nom} = 313.15$ K		
0.85	313.21	64.0	1315	64.0
3.25	313.23	65.6	1330	65.6
6.25	313.12	67.5	1347	67.5
9.12	313.15	69.2	1362	69.2
12.11	313.10	70.9	1376	70.9
15.20	313.04	72.6	1389	72.6
17.38	313.10	73.6	1398	73.6
		$T_{nom} = 333.15$ K		
1.30	333.10	58.4	1234	58.4
4.75	333.00	61.0	1265	61.0
8.23	332.95	63.4	1291	63.4
11.66	332.85	65.5	1312	65.4
15.24	332.99	67.6	1331	67.6
18.08	332.89	69.1	1345	69.0

Table III. Measurements of Thermal Conductivity of R125

Pressure P (MPa)	Temperature T (K)	Thermal conductivity $\lambda(T, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$	Density $\rho(T_{nom}, P)$ $(kg \cdot m^{-3})$	Thermal conductivity $\lambda(T_{nom}, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$
		$T_{nom} = 253.15$ K		
1.24	253.04	79.3	1412	79.3
		$T_{nom} = 273.15$ K		
1.25	273.11	71.3	1325	71.3
3.19	273.30	72.4	1338	72.4
5.19	273.21	73.6	1351	73.7
7.16	273.31	74.9	1362	75.0
9.20	273.34	76.3	1373	76.4
11.05	273.48	77.6	1382	77.7
		$T_{nom} = 293.15$ K		
1.25	293.20	62.9	1219	63.0
4.55	292.93	66.1	1257	66.0
7.15	292.94	68.2	1280	68.2
9.53	292.86	70.1	1298	70.0
11.61	293.08	71.5	1311	71.4
13.66	293.14	72.7	1323	72.7
16.03	293.13	74.1	1337	74.1
		$T_{nom} = 313.15$ K		
3.88	312.82	57.8	1134	57.6
5.70	312.88	59.8	1164	59.8
7.65	312.98	61.9	1189	61.8
9.58	313.04	63.7	1210	63.6
11.60	313.11	65.3	1229	65.4
13.50	313.46	66.7	1243	66.8

Figure 2 shows the deviations of other investigators' experimental values of the thermal conductivity of the four refrigerants along the saturation line, relative to the values calculated by Eq. (1). Only measurements performed over the last 4 years were considered. The measurements of the thermal conductivity of R32, R124, R125, and R141b of Yata et al. [17, 18] were performed at high pressures in a transient hot-wire instrument (two bare Pt wires) with no quoted uncertainty. The deviations of the extrapolated-to-saturation values are shown in Fig. 2. On average, these measurements deviate by 3% from the present values, but also show a systematically different temperature slope. This slightly different temperature slope, that does not seem to agree with most other sets of measurements, was also

Table IV. Measurements of Thermal Conductivity of R141b

Pressure P (MPa)	Temperature T (K)	Thermal conductivity $\lambda(T, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$	Density $\rho(T_{nom}, P)$ $(kg \cdot m^{-3})$	Thermal conductivity $\lambda(T_{nom}, P)$ $(mW \cdot m^{-1} \cdot K^{-1})$
		$T_{nom} = 253.15$ K		
0.30	253.25	104.3	1316	104.3
3.27	253.32	105.2	1320	105.2
6.24	253.29	106.1	1324	106.1
9.22	253.33	107.0	1327	107.0
11.02	253.32	107.5	1330	107.6
13.95	253.38	108.3	1333	108.4
		$T_{nom} = 273.15$ K		
0.27	273.31	98.5	1280	98.5
3.70	273.25	99.6	1286	99.7
7.19	273.28	100.8	1292	100.9
10.65	273.26	102.0	1297	102.0
14.34	273.30	103.2	1303	103.2
17.68	273.19	104.3	1309	104.3
20.93	273.22	105.3	1314	105.3
		$T_{nom} = 293.15$ K		
0.27	293.40	92.6	1244	92.7
3.71	293.31	93.9	1250	94.0
7.08	293.30	95.2	1257	95.3
10.63	293.43	96.5	1263	96.6
14.14	293.47	97.8	1270	97.8
17.65	293.44	99.0	1276	99.1
20.43	293.57	99.9	1281	100.0
		$T_{nom} = 313.15$ K		
0.41	313.21	86.9	1205	86.9
4.15	313.15	88.5	1214	88.5
7.72	313.17	90.0	1222	90.0
10.63	313.27	91.1	1228	91.1
14.25	313.23	92.5	1235	92.6
17.74	313.25	93.8	1241	93.9
21.71	313.33	95.3	1248	95.3
		$T_{nom} = 333.15$ K		
0.45	333.04	81.2	1165	81.1

Table V. Coefficients and Constants in Eq. (1)

	R32	R124	R125	R141b
Coefficients C_{ij} ($mW \cdot m^{-1} \cdot K^{-1}$)				
C_{00}	345.53	188.25	163.60	178.00
C_{01}	−219.23	−191.99	−82.956	−139.06
C_{02}	−56.202	43.383	−40.537	0
C_{10}	19.377	4.7622	−43.343	−0.906
C_{11}	−71.999	−13.051	−82.025	4.1378
C_{12}	69.981	12.872	−31.947	0
C_{20}	13.218	−0.6703	20.195	0.0811
C_{21}	−28.383	2.0400	−43.185	−0.1645
C_{22}	14.017	−1.6290	22.665	0
P_c (MPa)	5.793	3.66	3.631	4.230
T_c (K)	351.36	395.65	339.40	477.26
σ (%)	±0.06	±0.03	±0.05	±0.03

Table VI. The Thermal Conductivity of R32, R124, R125, and R141b Along the Saturation Line

Liquid	Temperature T_s (K)	Pressure P_s (MPa)	Density ρ_s ($kg \cdot m^{-3}$)	Thermal conductivity $\lambda(T_s, P_s)$ ($mW \cdot m^{-1} \cdot K^{-1}$)
R32	253.15	0.406	1121	158.7
	273.15	0.812	1055	141.9
	293.15	1.473	982	125.5
	313.15	2.478	893	110.0
R124	253.15	0.072	1484	83.5
	273.15	0.163	1430	76.5
	293.15	0.327	1377	70.0
	313.15	0.593	1313	63.9
	333.15	0.996	1231	58.2
R125	253.15	0.338	1407	79.2
	273.15	0.671	1320	71.0
	293.15	1.206	1219	62.9
	313.15	2.019	1089	55.3
R141b	253.15	0.010	1316	104.2
	273.15	0.028	1280	98.4
	293.15	0.065	1243	92.6
	313.15	0.133	1205	86.8
	333.15	0.246	1164	81.0

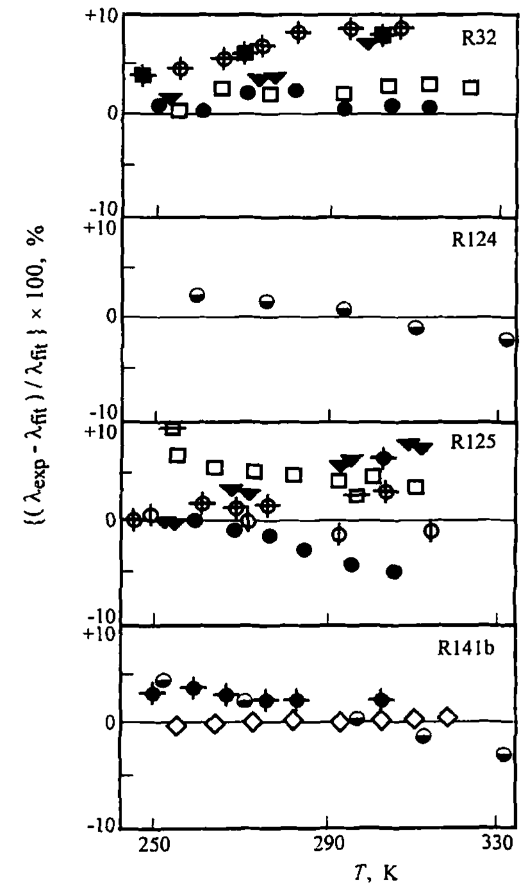

Fig. 2. Percentage deviations of the thermal-conductivity measurements of R32, R124, R125, and R141b along the saturation line, from Eq. (1). (●) Yata et al. [17]; (⊖) Yata et al. [18]; (□) Gross and Song [19]; (-⊕-) Papadaki and Wakeham [20]; (-◆-) Papadaki et al. [21]; (▼) Bivens et al. [22]; (-✱-) Ro et al. [23]; (Φ) Perkins [24]; (◇) Gurova et al. [25]; (-⊟-) Wilson et al. [26].

noticed in our other refrigerant measurements [2, 3]. Two bare Pt were also employed in the transient hot-wire instrument of Gross and Song [19] for the measurement of the thermal conductivity of R32 and R125. These investigators, moreover, employed the polarization-voltage technique and quote an uncertainty of $\pm 1.6\%$. The deviations of the extrapolated-to-saturation values, shown in the same figure, rise up to 7% in the case of R125. Papadaki et al. [20, 21] employed also a transient hot-wire instrument (two anodized Ta wires) for the measurement of the thermal conductivity of R32, R125, and R141b aong the saturation line. The quoted uncertainty was about ±1.5%. Although the measurements of R125 and R141b are just over the mutual uncertainty of the two instruments, the deviations of the R32 measurements rise up to 9%. This alarming difference is probably attributed to the purity of their sample, according to discussions between the two groups, as it was one of the first samples produced by their supplier.

The thermal conductivity of R32 and R125 was also measured along the saturation line by Bivens et al. [22] employed a transient hot-wire instrument (one bare Pt wire) on a relative basis, with a quoted uncertainty of ±1%. These measurements show deviations, from the values calculated by Eq. (1), of up to 9%, and a distinctively different temperature profile that seems to disagree with most other investigators.

No other researcher, to our knowledge, has recently measured the thermal conductivity of more than one of these refrigerants. The measurements of the thermal conductivity of R32 by Ro et al. [23], performed in a transient hot wire (one Pt wire) with a quoted uncertainty of ±1%, show deviations of up to 8% from the present set. This is both very alarming and unexplainable, as there is usually good agreement between the two groups. Perkins [4] and Gurova et al. [5] employed also a transient hot-wire instrument (two bare Pt wires) with the polarization technique, with quoted uncertainties of +1.0 and ±0.5%, respectively. The extrapolated-to-saturation values of R125 of Perkins [24] and the saturation values of R141b of Gurova et al. [5] are both in good agreement with the present values. Finally, the measurements of R125 of Wilson et al. [26], performed in a transient hot-wire instrument (one bare Pt wire) on a relative basis along the saturation line with a quoted uncertainty of ±1.5%, deviate by up to 10% from the present values.

Figures 3 to 6 show the deviations of the high-pressure measurements of the thermal conductivity of other investigators from the values calculated by Eq. (1). In the case of the thermal conductivity of R124 and R141b, at high pressures, only Yata et al. [18] have reported measurements. In the case of R32, high-pressure measurements have been reported by Yata et al. [17], Gross and Song [19], and Ro et al. [23]. Finally, high-pressure measurements of R125 have been reported by Yata et al. [17], Gross and Song [19], and Perkins [24]. It can be seen that all these of measurements follow the same pattern of deviations, as discussed above for the values along the saturation line.

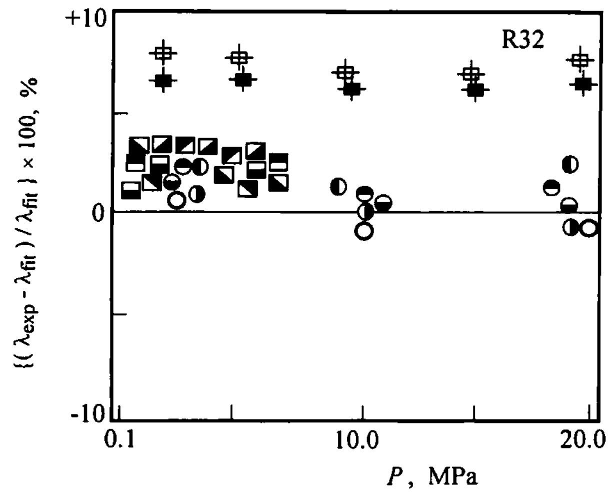

Fig. 3. Percentage deviations of the experimental high-pressure thermal-conductivity measurements of R32 from Eq. (1). Yata et al. [18]: (⊖) 253 K; (◓) 275 K; (◐) 284 K; (◑) 304 K; (○) 314 K. Gross and Song [19]: (⊟) 253 K; (⬓) 263 K; (◩) 273 K; (◪) 283 K; (◩) 303 K; (◪) 313 K. Ro et al. [23]: (✚) 273.15 K; (⊞) 298.15 K.

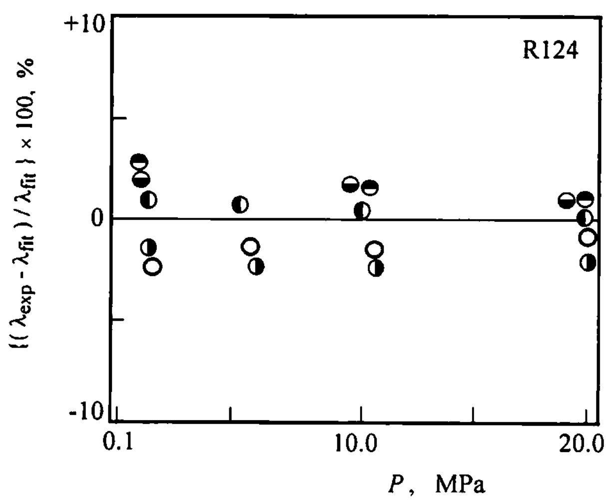

Fig. 4. Percentage deviations of the experimental high-pressure thermal-conductivity measurements of R124 from Eq. (1). Yata et al. [18]: (⊖) 257 K; (◓) 275 K; (◐) 295 K; (◑) 315 K; (○) 335 K.

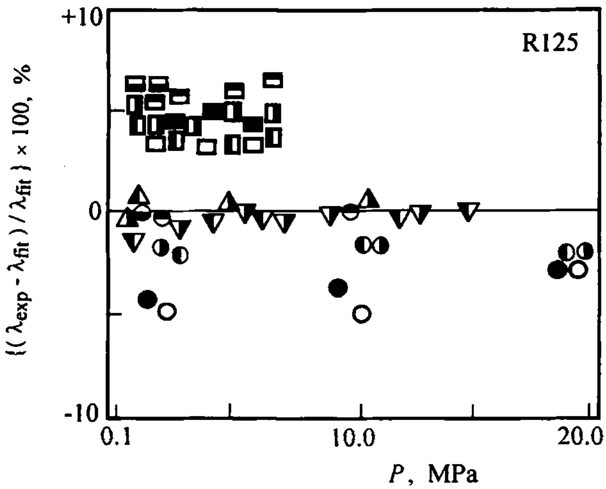

Fig. 5. Percentage deviations of the experimental high-pressure thermal-conductivity measurements of R125 from Eq. (1). Yata et al. [17]: (◒) 257 K; (◓) 267 K; (◑) 276 K; (◐) 285 K; (●) 295 K; (○) 305 K. Gross and Song [19]: (⬓) 253 K; (⬒) 263 K; (◨) 273 K; (◧) 283 K; (■) 303 K; (□) 313 K. Perkins [24]: (▲) 252 K; (△) 272 K; (▽) 292 K; (▼) 312 K.

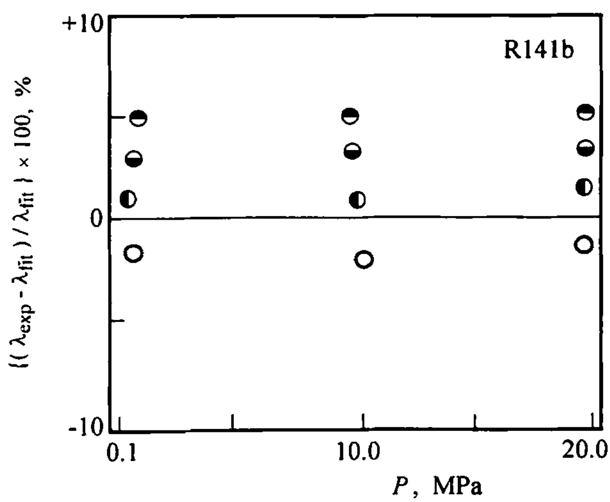

Fig. 6. Percentage deviations of the experimental high-pressure thermal-conductivity measurements of R141b from Eq. (1). Yata et al. [18]: (◓) 252 K; (◒) 273 K; (◐) 292 K; (○) 313 K.

4. CONCLUSION

Measurements of the thermal conductivity of R32, R124, R125, and R141b have been presented. The measurements, performed in a transient hot-wide instrument employing for the heat source two electrical-insulated tantalum wires, cover a temperature range from 253 to 334 K at pressures up to 20 MPa. The overall uncertainty in the reported data is $\pm 0.5\%$, an estimate confirmed by the measurement of the thermal conductivity of water and toluene. Certainly the general picture for thermal-conductivity measurements is slowly improving compared with the existing situation a few years ago. Nevertheless, it seems, at present, that the thermal conductivity of these four environmentally friendly refrigerants is known only with an uncertainty of about $\pm 5.0\%$. From the discussion of the results, it seems that still more measurements of high accuracy are required.

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