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## Ultrasonic Speeds in Liquid Monochlorodifluoromethane (R22) and Monochloropentafluoroethane (R115) under High Pressures

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The ultrasonic speeds in liquid monochlorodifluoromethane (R22) and monochloropentafluoroethane (R115) were measured by using a ring-around technique employing a fixed-path ultrasonic interferometer of 2 MHz. The results cover every 5 K in the range of temperatures from 283.15 to 323.15 K and pressures from near their saturated vapor pressures to about 50 MPa. The experimental uncertainty of ultrasonic speed was estimated to be no greater than  $\pm 0.34\%$  up to 10 MPa and  $\pm 0.23\%$  above 10 MPa. From the experimental results, the isentropic compressibility and the ratio of heat capacities were determined by using the  $pVT$  data reported elsewhere. The present results were compared with our previous results for R502, an azeotropic refrigerant mixture of R22 and R115.

### Introduction

In an earlier paper, we reported the temperature and pressure effects of the ultrasonic speed and the isentropic compressibility for compressed liquid R502 (1). This refrigerant is the azeotropic mixture of 48.8 wt % (63 mol %) monochlorodifluoromethane, R22, and 51.2 wt % (37 mol %) monochloropentafluoroethane, R115. For these refrigerants, the studies on experimental  $pVT$  and/or the formulation of an equation of state have been investigated in wide ranges of temperature and pressure (2, 3). However, the direct measurement on thermodynamic properties in connection with the variation due to pressure has scarcely been reported. In this paper, the ultrasonic speeds in the liquid phase for R22 and R115 were measured in the range of temperatures from 283.15 to 323.15 K and pressures from near the saturated vapor pressure to about 50 MPa. From the experimental speed, the isentropic compressibility and the ratio of heat capacities were determined by using the  $pVT$  data reported elsewhere. The temperature, pressure, and composition dependences of these quantities were examined in comparison with those of R502 observed in our recent work (1).

Table I. Physical Properties of Each Compound

	R22	R115	R502 <sup>a</sup>
chemical formula	CHClF <sub>2</sub>	CClF <sub>2</sub> -CF <sub>3</sub>	R22/R115
molecular weight	86.48	154.48	111.64
dipole moment, <sup>b</sup> 10 <sup>-30</sup> C·m	4.73	1.73	
critical constants <sup>c</sup>			
temp, K	369.15	353.15	355.35
press., MPa	4.98	3.23	4.08
density, kg·m <sup>-3</sup>	524	613	561

<sup>a</sup> Azeotropic mixture refrigerant. <sup>b</sup> Reference 4. <sup>c</sup> Reference 3.

### Experimental Section

**Material.** Monochlorodifluoromethane, CHClF<sub>2</sub> (R22), and monochloropentafluoroethane, CClF<sub>2</sub>-CF<sub>3</sub> (R115), were supplied by Daikin Kogyo Co. Their purities were better than 99.9 wt % as measured by GLC. The physical properties of each refrigerant are listed in Table I together with those of R502.

**Apparatus.** The method used for measurement of ultrasonic speed was a ring-around technique with fixed-path ultrasonic interferometer employing a single transducer, similar to that described previously (5). The measurements were covered in detail over the range of temperature from 283.15 to 323.15 K and pressures from near the saturated vapor pressure to about 50 MPa. The uncertainty in temperature measurements of the sample was less than  $\pm 0.03$  K. The uncertainties of pressure, measured by a precise bourdon gauge and a strain gauge, were estimated to be no greater than  $\pm 0.03$  MPa in the range up to 5 MPa and  $\pm 0.12$  MPa above 5 MPa to 50 MPa. The probable uncertainty due to the instrument used in this work was confirmed by measuring the speed in pure benzene to be less than  $\pm 1.3\%$  under all present experimental conditions (5).

### Results and Discussion

The experimental values of the ultrasonic speeds  $u$  in the liquid phase of monochlorodifluoromethane (R22) and monochloropentafluoroethane (R115) at several temperatures  $T$  and

**Table II. Ultrasonic Speed  $u$ , Density  $\rho$ , and Isentropic Compressibility  $\kappa_S$  at Various Temperatures  $T$  and Pressures  $p$** 

$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$
R22											
283.15 K											
0.680 <sup>b</sup>	622.3 <sup>c</sup>	1246.7 <sup>d</sup>	207.8	3.65	649.0	1259.2	188.5	30.69	820.6	1341.9	110.6
0.94	623.6	1247.9	206.0	4.10	652.8	1260.9	186.0	35.40	843.2	1352.9	103.9
1.01	625.4	1248.2	204.8	4.65	657.5	1263.1	183.1	39.53	862.1	1362.1	98.7
1.53	630.2	1250.4	201.3	8.36	686.3	1277.0	166.2	43.78	880.8	1371.0	94.0
1.93	633.7	1252.1	198.8	12.73	717.3	1291.8	150.4	47.55	895.1	1378.6	90.5
2.32	637.2	1253.7	196.4	17.31	746.2	1306.1	137.5	51.10	910.3	1385.4	87.1
2.73	641.1	1255.5	193.8	22.42	777.5	1320.7	125.2				
3.13	644.4	1257.1	191.5	26.72	800.2	1332.0	117.2				
288.15 K											
0.680 <sup>b</sup>	599.0 <sup>c</sup>	1228.6 <sup>d</sup>	226.8	4.20	630.6	1244.5	202.0	24.79	771.2	1314.5	127.9
0.90	600.2	1229.6	225.7	4.48	633.0	1245.7	200.3	29.08	794.2	1326.0	119.5
1.25	603.8	1231.3	222.7	7.06	654.4	1256.3	185.8	33.38	815.9	1336.7	112.4
2.54	615.6	1237.3	213.3	9.52	673.5	1265.8	174.1	37.43	835.1	1346.4	106.5
3.10	620.5	1239.7	209.4	13.32	700.9	1279.3	159.1	40.94	850.9	1354.3	101.9
3.47	624.1	1241.4	206.8	16.44	721.5	1286.6	148.9	45.02	869.1	1363.1	97.1
3.73	626.4	1242.5	205.1	20.32	745.5	1301.7	138.2	49.90	889.0	1373.1	92.1
293.15 K											
0.910 <sup>b</sup>	574.5 <sup>c</sup>	1209.9 <sup>d</sup>	250.3	3.20	597.8	1221.3	229.1	24.21	751.9	1298.6	136.2
0.95	574.9	1210.2	250.0	3.65	602.3	1223.4	225.2	28.22	774.4	1309.9	127.2
1.17	577.1	1211.3	247.9	4.13	606.8	1225.7	221.5	30.88	788.5	1317.0	122.1
1.45	579.8	1212.7	245.2	7.36	635.5	1239.9	199.6	34.21	805.5	1325.6	116.2
1.64	582.1	1213.7	243.1	10.37	659.9	1252.1	183.4	37.55	821.9	1333.8	110.9
2.11	586.8	1216.0	238.7	13.71	684.5	1264.6	168.7	40.53	835.8	1340.7	106.7
2.30	588.9	1217.0	236.9	16.91	706.7	1275.7	156.9	43.23	848.2	1346.8	103.1
2.84	594.2	1219.6	232.1	21.05	732.4	1289.1	144.6	50.71	880.2	1362.8	94.7
298.15 K											
1.049 <sup>b</sup>	550.0 <sup>c</sup>	1190.7 <sup>d</sup>	277.5	4.01	581.9	1206.6	244.7	27.37	752.8	1294.9	136.2
1.17	550.7	1191.4	276.8	4.35	585.4	1208.3	241.4	31.18	774.1	1305.7	127.8
1.35	553.1	1192.4	274.1	4.81	589.9	1210.6	237.3	36.38	799.4	1319.4	118.5
1.81	558.4	1195.0	268.4	7.05	610.3	1221.3	219.8	39.81	816.1	1327.9	113.0
1.98	560.4	1195.9	266.2	10.37	639.4	1236.0	197.9	43.07	832.7	1335.6	107.9
2.33	564.3	1197.8	262.1	13.17	661.2	1247.3	183.3	47.33	851.6	1345.2	102.4
2.72	568.0	1199.9	258.2	16.51	685.5	1259.8	168.9	50.90	866.8	1352.9	98.3
2.92	570.6	1201.0	255.7	20.05	709.2	1272.1	156.3				
3.44	576.1	1203.7	250.3	23.38	729.3	1282.6	146.5				
303.15 K											
1.196 <sup>b</sup>	525.1 <sup>c</sup>	1170.8 <sup>d</sup>	309.6	4.15	559.2	1188.1	269.1	29.17	745.6	1286.5	139.8
1.38	526.9	1171.9	307.3	4.65	564.4	1190.8	263.6	31.91	761.8	1294.4	133.1
1.58	529.5	1173.2	303.9	7.72	594.2	1206.5	234.7	35.41	780.4	1304.0	125.9
1.83	532.4	1174.7	300.3	10.05	614.8	1217.4	217.3	39.16	799.1	1313.7	119.2
2.18	536.7	1176.8	294.9	13.09	639.9	1230.6	198.4	42.81	815.1	1322.7	113.7
2.64	542.0	1179.5	288.6	16.46	665.2	1244.0	181.6	47.23	835.2	1333.1	107.5
3.02	546.5	1181.7	283.2	21.04	696.6	1260.6	163.4	51.05	851.7	1341.6	102.7
3.44	551.2	1184.1	277.9	24.95	721.0	1273.5	151.0				
308.15 K											
0.788 <sup>b</sup>	494.0 <sup>c</sup>	1150.2 <sup>d</sup>	356.2	4.64	541.2	1175.3	290.4	26.74	715.7	1270.5	153.6
1.23	499.1	1153.4	347.9	7.18	567.7	1189.8	260.7	30.84	739.4	1283.4	142.5
1.83	507.1	1157.4	336.0	10.04	594.9	1204.5	234.5	33.15	752.0	1290.2	137.0
2.43	514.8	1161.5	324.8	13.52	624.5	1220.7	210.0	37.67	775.8	1303.0	127.5
2.96	521.4	1165.0	315.6	16.21	645.5	1232.1	194.7	41.68	795.7	1313.5	120.4
3.55	528.6	1168.7	306.2	19.86	671.8	1246.5	177.7	45.63	814.3	1323.4	113.9
3.99	533.6	1171.4	299.8	23.08	693.0	1258.2	165.4	50.19	834.5	1334.1	107.6
313.15 K											
1.529 <sup>b</sup>	475.2 <sup>c</sup>	1128.6 <sup>d</sup>	392.2	4.32	512.6	1148.3	331.3	25.93	694.5	1248.3	166.0
1.90	480.0	1131.4	383.5	4.79	518.4	1151.4	323.1	29.70	717.4	1260.8	154.1
2.28	485.6	1134.2	373.8	6.63	538.8	1162.7	296.2	34.09	742.2	1274.3	142.4
2.57	489.2	1136.3	367.7	10.25	575.5	1182.8	255.2	37.79	762.0	1284.9	134.0
3.11	496.3	1140.1	356.0	12.82	598.9	1195.7	233.1	41.38	780.2	1294.7	126.8
3.50	501.9	1142.8	347.3	17.24	634.8	1215.4	204.1	45.89	802.0	1306.2	119.0
3.87	506.5	1145.3	340.2	22.10	669.6	1234.6	180.6	50.66	823.5	1317.7	111.8
318.15 K											
1.726 <sup>b</sup>	451.7 <sup>c</sup>	1106.2 <sup>d</sup>	442.9	6.46	515.0	1141.0	330.3	26.84	684.9	1236.0	172.4
2.09	456.5	1109.2	432.6	9.60	549.4	1160.0	285.5	29.91	703.7	1246.5	162.0
2.65	464.9	1113.7	415.3	9.75	552.2	1160.9	282.4	34.25	728.0	1260.3	149.7
3.01	470.5	1116.5	404.5	12.62	579.6	1176.3	253.0	39.29	755.6	1275.0	137.3
3.60	479.0	1121.0	388.6	15.62	604.6	1190.9	229.7	42.31	770.8	1283.4	131.1
4.15	486.6	1125.1	375.3	18.76	629.1	1204.8	209.7	46.60	791.6	1294.6	123.2
4.77	493.6	1129.6	363.3	22.42	655.6	1219.7	190.7	51.28	812.9	1306.1	115.8

Table II (Continued)

$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$
323.15 K											
1.941 <sup>b</sup>	424.5 <sup>c</sup>	1082.4 <sup>d</sup>	512.6	7.19	499.9	1123.9	356.0	30.38	690.9	1232.0	169.9
2.11	426.0	1084.0	508.1	8.61	516.9	1133.2	330.2	34.33	714.2	1245.0	157.4
2.35	430.4	1086.2	496.9	11.05	543.0	1148.0	295.4	38.03	734.5	1256.3	147.5
2.77	437.6	1089.9	479.1	13.26	564.5	1160.3	270.4	41.48	752.5	1266.3	139.4
3.24	445.3	1093.9	461.0	17.25	599.2	1180.1	236.0	45.64	773.2	1277.6	130.9
3.84	454.9	1099.0	439.6	20.57	625.2	1194.8	214.1	50.02	793.9	1288.9	123.0
4.20	460.2	1101.8	428.5	24.43	652.6	1210.5	193.9				
4.93	470.6	1107.6	407.6	27.32	672.0	1221.3	181.3				
R115											
283.15 K											
0.600 <sup>b</sup>	414.1 <sup>c</sup>	1369.5 <sup>d</sup>	360.6	3.96	453.1	1393.7	349.4	22.85	601.9	1495.4	184.5
0.97	417.9	1369.4	418.0	4.73	461.3	1399.4	335.8	26.95	626.2	1511.3	168.7
1.01	418.5	1369.7	416.5	6.73	481.0	1413.2	305.7	32.11	654.8	1529.5	152.4
1.75	427.8	1376.1	397.0	9.72	508.1	1431.8	270.5	36.17	675.0	1542.7	142.2
2.30	434.5	1380.7	383.6	13.75	540.2	1454.0	235.6	41.46	700.2	1558.6	130.8
2.94	441.9	1385.8	369.5	16.14	557.8	1465.9	219.2	44.72	715.1	1567.8	124.7
3.63	449.5	1391.2	355.7	19.04	577.6	1479.3	202.5	48.84	733.1	1578.8	117.8
288.15 K											
0.692 <sup>b</sup>	394.0 <sup>c</sup>	1332.5 <sup>d</sup>	483.2	4.24	436.2	1363.6	385.2	29.18	625.6	1492.7	171.1
1.06	397.9	1336.0	472.7	4.89	442.5	1368.7	373.0	33.67	649.8	1508.4	156.9
1.72	407.5	1342.2	448.5	6.58	461.5	1381.1	339.9	37.99	671.6	1522.3	145.6
2.16	413.6	1346.2	434.1	8.93	484.6	1396.9	304.7	42.18	691.8	1535.0	136.1
2.63	417.1	1350.3	425.6	12.38	514.1	1417.5	266.8	47.12	714.1	1548.9	126.6
2.67	418.7	1350.6	422.2	17.61	553.3	1444.6	226.1	51.34	732.3	1560.1	119.5
3.22	423.9	1355.3	410.4	22.24	584.1	1465.4	199.9				
3.61	428.5	1358.6	400.8	25.92	606.7	1480.4	183.4				
293.15 K											
0.801 <sup>b</sup>	373.1 <sup>c</sup>	1309.5 <sup>d</sup>	548.5	4.54	421.6	1345.2	418.1	27.93	605.9	1474.1	184.7
0.96	374.4	1311.2	543.9	7.15	450.1	1365.8	361.2	31.94	628.4	1489.0	170.7
1.61	383.9	1318.0	514.7	10.29	480.3	1387.5	312.3	36.88	654.3	1505.9	155.1
2.19	391.8	1323.8	491.9	14.26	513.8	1411.2	268.4	40.73	673.1	1518.0	145.3
2.57	397.0	1327.5	478.0	18.34	544.5	1432.5	235.4	46.15	698.5	1534.0	133.6
3.16	404.8	1333.0	457.7	22.11	570.7	1450.0	211.7	48.67	709.6	1541.0	128.8
3.95	414.6	1340.2	433.9	23.86	581.3	1457.6	202.9	49.04	711.0	1542.0	128.2
298.15 K											
0.911 <sup>b</sup>	352.0 <sup>c</sup>	1290.8 <sup>d</sup>	624.9	8.62	449.0	1363.7	363.7	32.65	620.2	1486.4	174.9
0.97	351.6	1291.5	626.1	12.91	488.2	1393.0	301.1	37.71	646.7	1504.3	158.9
1.61	362.0	1299.1	587.1	16.25	515.1	1412.7	266.7	41.28	664.7	1516.1	149.2
2.07	369.1	1304.4	562.5	20.37	545.2	1434.3	234.4	44.32	679.1	1525.5	142.1
2.51	376.0	1309.2	540.2	23.60	566.8	1449.5	214.7	46.14	687.5	1531.0	138.2
3.07	383.6	1315.1	516.1	28.33	595.9	1469.7	191.5	49.65	703.0	1541.1	131.2
4.68	404.2	1330.9	459.7	28.94	599.4	1472.2	189.0				
303.15 K											
1.039 <sup>b</sup>	329.9 <sup>c</sup>	1260.6 <sup>d</sup>	722.8	4.67	384.0	1302.3	520.7	26.82	574.4	1439.5	210.5
1.19	330.8	1262.6	723.5	6.53	408.4	1319.7	454.3	30.75	597.7	1455.4	192.2
1.65	339.1	1268.6	685.5	9.30	438.9	1342.4	386.7	34.66	619.3	1470.0	177.3
2.08	346.4	1273.9	654.1	14.11	484.1	1375.2	310.2	38.96	641.9	1484.8	163.4
2.51	353.4	1279.1	625.8	17.84	513.2	1396.7	271.8	40.99	652.2	1491.4	157.6
3.03	361.4	1285.1	595.5	18.92	521.2	1402.5	262.4	42.65	660.4	1496.7	153.2
3.50	368.3	1290.2	571.1	20.84	535.1	1412.2	247.3	49.16	690.5	1516.0	138.3
4.08	376.5	1296.3	544.2	23.04	550.2	1422.8	232.1				
308.15 K											
1.176 <sup>b</sup>	309.2 <sup>c</sup>	1234.4 <sup>d</sup>	847.0	4.51	364.2	1278.8	589.5	30.13	582.4	1442.9	204.2
1.62	316.4	1241.3	804.5	6.86	394.7	1303.3	492.5	34.59	607.8	1460.4	185.3
2.09	325.1	1248.1	758.0	11.03	440.2	1338.8	385.3	43.94	655.9	1492.5	155.7
2.60	334.2	1255.4	713.0	17.19	494.5	1379.8	296.3	48.62	677.8	1506.8	144.4
3.09	342.4	1261.7	676.0	22.26	532.2	1407.3	250.8	49.66	682.6	1509.9	142.1
3.72	352.5	1269.5	633.9	25.34	552.8	1422.1	230.0				
313.15 K											
1.333 <sup>b</sup>	291.2 <sup>c</sup>	1206.6 <sup>d</sup>	977.4	10.00	414.3	1313.2	443.5	38.41	617.5	1464.7	179.0
2.24	305.8	1222.6	874.2	13.83	542.3	1343.3	363.8	40.14	626.5	1470.9	173.2
2.71	314.1	1230.2	823.6	17.22	481.5	1366.0	315.6	42.84	640.2	1480.1	164.8
3.22	323.7	1237.8	770.8	20.25	505.1	1383.9	283.1	44.47	648.2	1485.5	160.1
3.80	333.6	1246.2	720.8	26.67	549.4	1416.7	233.8	47.79	663.8	1496.0	151.6
4.69	346.8	1257.9	660.8	30.55	573.6	1434.0	211.9	50.88	677.7	1505.3	144.6
7.70	388.0	1291.7	514.2	34.78	598.0	1451.1	192.6				
318.15 K											
1.490 <sup>b</sup>	283.1 <sup>c</sup>	1176.9 <sup>d</sup>	1138.6	4.47	324.4	1231.8	771.0	9.64	394.7	1293.9	469.1
3.59	309.0	1217.9	859.5	4.75	329.2	1236.0	746.2	13.45	434.4	1327.3	399.1
3.93	315.1	1223.5	822.8	6.68	357.5	1261.7	620.0	16.50	462.2	1349.7	346.7

Table II (Continued)

$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$	$p/\text{MPa}$	$u/\text{m}\cdot\text{s}^{-1}$	$\rho^a/\text{kg}\cdot\text{m}^{-3}$	$\kappa_S/\text{TPa}^{-1}$
20.12	491.6	1372.9	301.4	29.47	555.9	1421.7	227.6	39.84	614.6	1464.1	180.8
22.29	507.9	1385.5	279.8	31.20	566.4	1429.5	218.0	43.44	633.1	1476.9	168.9
25.91	533.0	1404.6	250.5	34.54	585.8	1443.6	201.8	48.37	656.6	1493.3	155.3
27.83	545.6	1414.0	237.5	37.78	603.6	1456.4	188.4				
323.15 K											
1.676 <sup>b</sup>	255.2 <sup>c</sup>	1146.2 <sup>d</sup>	1338.6	7.73	355.3	1259.5	628.7	27.08	529.5	1406.8	253.5
4.60	306.7	1212.9	875.9	10.91	384.8	1286.4	524.8	33.32	568.1	1436.2	215.7
4.75	309.2	1215.6	860.1	13.91	425.6	1322.3	417.5	38.63	597.9	1458.1	191.8
5.42	321.0	1226.8	791.0	16.35	448.2	1341.5	371.0	41.72	614.3	1469.8	180.2
6.21	333.6	1238.8	725.1	18.92	469.9	1359.5	333.0	45.54	633.4	1483.3	168.0
6.39	336.2	1241.4	712.5	24.10	509.0	1391.0	277.4	50.09	654.9	1498.4	155.5

<sup>a</sup> References 8 and 9. <sup>b</sup>  $p_s$ . <sup>c</sup>  $u_{p_s}$ . <sup>d</sup> Reference 10.

Table III. Coefficients  $a_i$  of Eq 1, Average Deviation  $\delta_{av}$ , and Maximum Deviation  $\delta_{max}$ <sup>a</sup>

$T/\text{K}$	$a_0$	$a_1$	$-a_2$	$10^3 a_3$	$-10^6 a_4$	$\delta_{av}/\%$	$\delta_{max}/\%$
R22							
283.15	615.98	9.437	0.1366	1.872	1.195	0.008	0.19
288.15	592.59	9.573	0.1286	1.558	0.874	0.064	0.17
293.15	564.82	10.835	0.1884	2.961	2.055	0.004	0.08
298.15	538.08	11.631	0.2087	3.097	1.941	0.002	0.08
303.15	510.59	12.471	0.2434	3.995	2.846	0.008	0.13
308.15	483.56	13.503	0.2828	4.697	3.286	0.043	0.13
313.15	453.24	14.913	0.3419	5.925	4.224	0.042	0.15
318.15	425.12	16.076	0.3800	6.453	4.451	0.084	0.16
323.15	391.23	18.029	0.4725	8.580	6.252	0.010	0.29
R115							
283.15	406.55	12.647	0.2665	4.571	3.312	0.065	0.14
288.15	385.14	13.098	0.2671	4.272	2.872	0.117	0.34
293.15	361.69	14.496	0.3417	6.082	4.449	0.084	0.17
298.15	338.10	15.709	0.3955	7.184	5.272	0.118	0.22
303.15	312.37	17.337	0.4799	9.182	6.947	0.122	0.46
308.15	287.73	18.947	0.5621	11.057	8.470	0.228	0.42
313.15	266.35	19.347	0.5467	10.183	7.463	0.198	0.33
318.15	244.39	20.144	0.5719	10.696	7.946	0.142	0.29
323.15	220.96	21.489	0.6300	11.788	8.640	0.166	0.25

<sup>a</sup>  $\delta_{av} = \{100[(u_{\text{exptl}} - u_{\text{calcd}})/u_{\text{calcd}}]\}/n$ ;  $n$  is the number of points.

pressures  $p$  are presented in Table II. The thermodynamic properties of saturated vapor pressure have particular importance in the case of fluorocarbon refrigerants, as a measure for learning the characteristics of each fluid. The measurements of the speed presented in this work were carried out at narrow pressure intervals near the saturation line in order to determine the ultrasonic speed for saturated liquid by the extrapolation of the data at high pressure. The  $u$  values for each isotherm increase smoothly with increasing pressure as illustrated graphically in Figure 1. The variations of experimental ultrasonic speed with pressure are represented by the polynomial equation

$$u/(\text{m}\cdot\text{s}^{-1}) = \sum_{i=0}^4 a_i (\rho/\text{MPa})^i \quad (1)$$

where  $a_i$  are coefficients for individual isotherms. The values of the coefficient  $a_i$  for both refrigerants, obtained by least-squares analysis of the experimental results, are listed in Table III together with the average and maximum deviations. The solid lines indicated in Figure 1 were calculated from the above equation. The large deviations from the curve were observed chiefly in lower pressure region, where there is a strong pressure effect on speed. In the present work, the ultrasonic speed was obtained by measuring the traversing time of a short acoustic pulse between a transducer and refractor. However, in the vicinity of vapor pressure the absorption of the acoustic wave in the sample occurs frequently ( $\delta$ ) and that increases with increasing temperature. This phenomenon, which appeared as a narrow pulse width in the received signal, gives rise to irregularity of the experimental speed. From these facts, the

probable uncertainty of the present values was  $\pm 0.34\%$  in the range up to 10 MPa and  $\pm 0.23\%$  above 10 MPa, taking into account the observed errors of temperature and pressure.

From the coefficients  $a_i$  for eq 1, the ultrasonic speed in the liquid phase,  $u_{p_s}$ , under the vapor pressure,  $p_s$ , was determined, and the results are also listed in Table II with  $p_s$  derived from the equation reported in ref 3. The relationships between the  $u_{p_s}$  values and temperature are shown in Figure 2 with those for R502. The curve for each refrigerant is a nearly straight line throughout the range of experimental conditions. It is estimated that these extrapolated values have maximum errors of  $\pm 0.6\%$  for R22 and  $\pm 0.8\%$  for R115 caused by the absorption of acoustic waves, which becomes enhanced as it reaches the critical point. For R22 and R115, Kokernak and Feldman (7) estimated the speed of sound for saturated liquids by a theoretical rule. The value at 298.15 K reported in their paper was  $472.4 \text{ m}\cdot\text{s}^{-1}$  for R22 and  $304.8 \text{ m}\cdot\text{s}^{-1}$  for R115. However, these values were ascertained to differ by about 14% for both refrigerants from those presented here. The present values may be considered more reliable than those estimated by Kokernak and Feldman because the measurements in this work were carried out accurately and in detail in the vicinity of the vapor pressure.

From the ultrasonic speed, the isentropic compressibility  $\kappa_T$ , which is very difficult to measure directly, can be obtained by the equation

$$\kappa = (\rho u^2)^{-1} \quad (2)$$

where  $\rho$  is density. Moreover, by combining the isothermal compressibility  $\kappa_T$  [ $= -1/V(\partial V/\partial p)_T$ ;  $V$ , specific volume] the

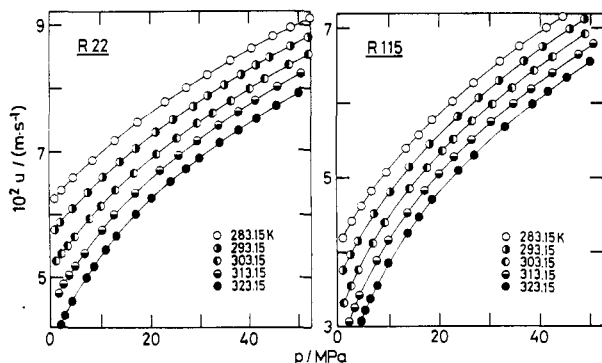


Figure 1. Pressure dependence of ultrasonic speed  $u$  in the liquid phase of the refrigerants R22 and R115.

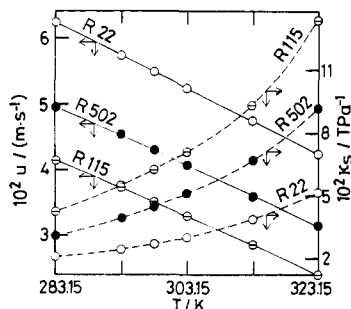


Figure 2. Temperature dependence of ultrasonic speed  $u$  and isentropic compressibility  $\kappa_S$  at the saturated liquid phase of the refrigerants R22, R115, and R502.

ratio of heat capacities  $\gamma$  at constant pressure  $C_p$  and constant volume  $C_v$  is given by

$$\gamma = C_p / C_v = \kappa_T \rho u^2 \quad (3)$$

The  $pVT$  properties in the liquid phase for R22 have been reported by Zander (2) and Kumagai and Iwasaki (8). However, very few data are available for R115. Kumagai and Iwasaki measured the  $pVT$  for R22 at temperatures every 20 K from 253.15 to 313.15 K at pressures up to 150 MPa with an accuracy better than  $\pm 0.13\%$ . For R115 Arakawa et al. (9) measured  $pVT$  data on five isotherms from 283.15 to 373.15 K and up to 100 MPa within  $\pm 0.09\%$ . These experimental values at each temperature were represented by the Tait equation. The density,  $\rho$ , and isothermal compressibility,  $\kappa_T$ , at arbitrary conditions, required in the calculation of  $\kappa_S$  and  $\gamma$ , were estimated, as each Tait parameter is shown with temperature. Also the density,  $\rho_p$ , at  $p_s$  was adapted from the data given by Okada (9).

The results of isentropic compressibility,  $\kappa_S$ , are also listed in Table II together with the densities corresponding to each condition of the speed measurement. Figure 3 presents the pressure dependence of  $\kappa_S$  on isotherms with the values for R502. The  $\kappa_S$  values for the high temperatures show a strong pressure dependence, especially those in the lower pressure region. Moreover, it is found that the results at the saturated vapor or its vicinity are influenced largely by the temperature change, as shown in Figure 3. On the other hand, when each value for the three refrigerants at constant temperature and pressure is displayed as a function of the mole fraction of R115, the magnitude for  $\kappa_S$  is found to increase in the order R22 < R502 < R115. This order produces a concave curve unlike that for the ultrasonic speed.

Next, the derived values of the ratio of heat capacities,  $\gamma$ , are given in Figure 4 as a function of pressure. For the lower temperature region the results decrease linearly with increasing pressure. However, the value at 323.15 K shows remarkable

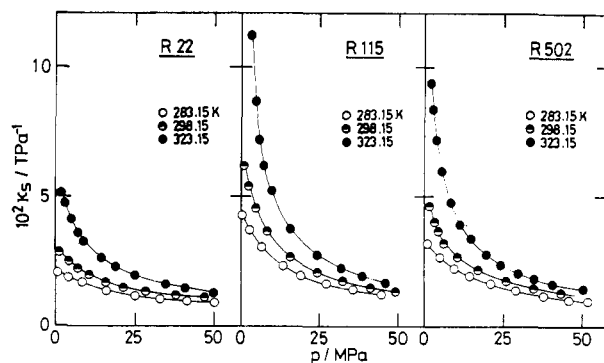


Figure 3. Pressure dependence of isentropic compressibility  $\kappa_S$  in the liquid phase of the refrigerants R22, R115, and R502.

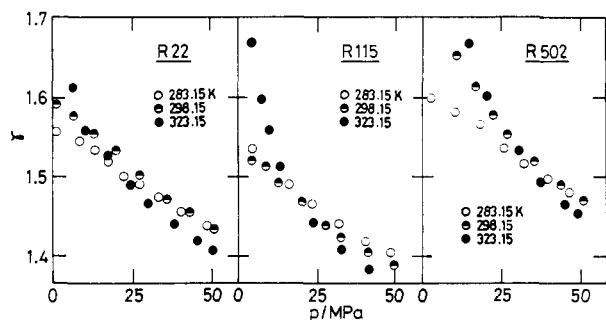


Figure 4. Pressure dependence of ratio of heat capacities  $\gamma$  in the liquid phase of the refrigerants R22, R115, and R502.

behavior as to pressure and temperature effects. The values, which are 1.67 for R22, 1.98 for R115, and 2.02 for R502 at vapor pressure, decrease indicating a strong pressure dependence, and in the higher pressure side they exhibit the lower values than those at low temperature. In our previous work on  $\gamma$  for organic liquids, (10, 11) the values for the nearly same condition, obtained from the velocity of sound, decreased with rise of pressure, but they did not show the large temperature or pressure effects and the behavior in curves crossed from the each other as illustrated in Figure 4. It is suggested that the pressure change of  $\gamma$  described above is the phenomenon characteristic of refrigerants near the critical condition, the values, as is usual, pass through infinity at the critical point. From these facts, the pressure and temperature effects on the ultrasonic speed, which is closely related to the several thermodynamic properties, near the critical condition are particularly interesting, and therefore detailed measurements in refrigerants near the critical temperature are expected in future.

Registry No. R 22, 75-45-6; R 115, 76-15-3.

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