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Speed of Sound in Binary Mixtures of Pentafluoroethane and 1,1-Difluoroethane from 243.15 K to 333.15 K and Pressures up to 30 MPa

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The speed of sound in the liquid phase for binary mixtures of pentafluoroethane (CHF_2CF_3) and 1,1-difluoroethane (CHF_2CH_3) was measured along six isotherms from (243 to 333) K and at pressures from near the saturation line up to about 30 MPa. The measurements were carried out by a sing-around technique operated at a frequency of 2 MHz employing the fixed-path acoustic interferometer. The combined uncertainty is estimated to be within ±0.2% in the high-density region. The speed of sound in the saturated liquid was estimated by an extrapolation of data obtained for the compressed liquid to the vapor pressure. The results for $(1 - x)CHF_2CF_3 + xCHF_2CH_3$ measured for mole fractions $x = 0.3806$, 0.6445, and 0.8447 were correlated by the polynomial equation as functions of temperature and pressure. The variations in the speed of sound with the composition at various temperatures and pressures are discussed for the system investigated as well as for $(1 - x)CHF_2CH_3 + xCF_3CH_2F$ and $(1 - x)CHF_2CH_3 + xCF_3CH_3$ reported elsewhere.

Introduction

In our previous papers, we have reported the speed of sound in dense liquids and the vapor pressure and/or bubble-point pressure for several pure hydrofluorocarbons (CHF_2CF_3,[1] CF_3CH_2F,[2] CF_3CH_3,[3] and CH_3CHF_2 [4]) and their mixtures ($CHF_2CF_3 + CF_3CH_2F$ [5] and $CHF_2CF_3 + CF_3CH_3$ [6]) measured over wide temperature and pressure ranges. These results give valuable information that can be used to investigate the thermophysical characterization of each pure substance and the resulting mixtures. In this work, which is a contribution to our research series on hydrofluorocarbons, the speed of sound in the liquid phase for the binary mixture of pentafluoroethane (CHF_2CF_3) and 1,1-difluoroethane (CH_3CHF_2) was measured from (243 to 333) K and at pressures up to about 30 MPa. The dependences of the speed of sound on temperature, pressure, and composition are discussed and compared with the data for other binary mixtures reported elsewhere.

Experimental Section

Pure samples of pentafluoroethane (CHF_2CF_3) and 1,1-difluoroethane (CH_3CHF_2) were supplied by Asahi Glass Co. Ltd. The substances were used without further purification except for careful drying with molecular sieves (4 Å, $^1/_{16}$, supplied by Wako Pure Chemicals Ind. Ltd.). The purities, checked by GLC, were found to be better than 99.96 mol %. The mixtures were prepared by mass with an uncertainty in mole fraction of ±0.0003.

The ultrasonic speeds were measured using the sing-around technique employing a fixed-path acoustic interferometer operated at a frequency of 2 MHz, similar to that described previously.[1] The pressure vessel containing the acoustic interferometer was immersed in a liquid thermostat filled with a mixture of ethylene glycol + water and controlled to within ±20 mK. The temperature was measured by a quartz thermometer, which was calibrated to within ±5 mK using a standard platinum thermometer (ITS-90). Pressure was observed by two precision strain gauges (Nagano Keiki Co., KH15) capable of measuring pressure to (5 ± 0.003) MPa, calibrated by a quartz crystal pressure transducer (Paroscientific Inc., 730-31K-101) and (35 ± 0.005) MPa, calibrated by a precision manometer (Tsukasa Sokken Co., PH-22-G). The speed of sound u [$= 2L/(t_2 - t_1)$] was obtained by measuring the period between the first, t_1, and second, t_2, echoes of a short acoustic pulse traveling a known distance, L [(23.801 ± 0.002) mm], at 298.15 K and 0.1 MPa between the transducer and reflector. The value of L was determined by measuring the difference $(t_2 - t_1)$ in liquid CCl_4 using the value of the speed of sound reported by Tamura et al.:[7] 921.11 $m \cdot s^{-1}$ at 298.15 K and 0.1 MPa. The difference $(t_2 - t_1)$ for the mixture investigated was from (51 to 102) μs and was recorded by a universal counter with a resolution of 0.1 ns as the average value of 1000 periods.

Results and Discussion

Experimental values of the speed of sound u in the liquid phase for the binary mixture of $(1 - x)CHF_2CF_3 + xCHF_2CH_3$ measured for three mole fractions $x = 0.3806$, 0.6445, and 0.8447 corresponding to about 75/25, 50/50, and 25/75 (CHF_2CF_3/CHF_2CH_3) mass ratios are listed in Table 1. Figure 1 presents the pressure dependences of the speed of sound for the mixture with $x = 0.6445$. At constant temperature, the speed of sound increases monotonically with increasing pressure; the largest pressure effect (i.e.,

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Table 1. Experimental Values of the Speed of Sound, u, in the Liquid Phase of $(1 - x)CHF_2CF_3 + xCHF_2CH_3$ at Various Mole Fractions, x, Temperature, T, and Pressure, p

p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{1})	p/MPa	u/(m·s^{-1})	p/MPa p	u/(m·s^{-1})
$x = 0.3806$															
$T = 243.15$ K															
0.195[a]	690.2[b]	2.504	706.8	4.792	723.2	7.670	742.3	11.210	764.5	16.045	792.8	22.539	827.6	27.580	852.8
0.849	694.6	3.065	710.8	5.795	730.0	9.360	753.3	13.082	775.7	17.589	801.3	24.924	839.7	28.855	858.8
1.143	696.8	3.853	716.4	6.848	737.0	9.798	755.8	14.603	784.5	20.328	816.1	26.426	847.1	30.081	864.6
2.057	703.5	4.353	719.9												
$T = 263.15$ K															
0.386[a]	596.6[b]	2.085	611.5	3.948	627.7	7.113	653.6	10.463	678.4	14.962	709.0	19.506	737.4	25.169	769.9
0.731	599.0	2.569	615.8	5.047	637.2	8.109	661.2	11.784	687.7	16.234	717.2	20.822	745.1	27.432	782.1
1.346	604.7	3.223	621.5	6.407	648.1	9.270	669.8	13.475	699.2	17.843	728.4	22.909	757.2	31.025	800.8
$T = 283.15$ K															
0.706[a]	502.2[b]	2.739	524.4	5.010	549.1	7.980	577.3	11.656	608.7	16.233	643.6	21.023	676.5	28.176	720.4
1.165	506.2	3.671	534.6	6.095	559.8	9.001	586.3	13.069	619.9	19.606	667.1	23.040	689.2	30.688	734.7
2.084	517.0	4.734	545.7	7.077	569.1	10.748	600.9	14.456	630.5	17.920	655.6	25.292	703.4		
$T = 298.15$ K															
1.082[a]	430.0[b]	3.231	459.4	4.957	482.1	7.987	516.0	11.638	551.6	16.078	589.4	19.527	615.8	25.425	656.5
1.751	439.5	3.779	466.7	5.895	493.2	8.969	526.1	13.410	567.3	16.113	589.7	21.216	627.9	27.784	671.5
2.398	448.6	4.476	475.5	6.805	503.4	10.058	536.8	14.591	577.3	17.471	600.3	23.675	645.0	29.753	683.6
2.750	452.9														
$T = 313.15$ K															
1.615[a]	356.2[b]	6.939	441.9	10.234	481.1	13.375	513.3	15.914	536.8	19.343	565.7	22.891	593.0	27.167	623.0
4.630	408.2	8.402	460.1	11.441	494.0	14.531	524.3	17.496	550.6	20.693	576.5	25.423	611.1	30.606	645.4
5.78	426.2	9.082	468.1												
$T = 333.15$ K															
2.525[a]	276.8[b]	7.777	372.1	10.088	405.8	13.295	445.3	16.159	475.7	19.293	505.4	22.481	532.7	28.145	575.6
6.951	358.4	8.980	390.4	11.575	425.0	14.976	463.6	17.408	487.9	20.699	517.8	24.872	551.7	30.230	590.4
$x = 0.6445$															
$T = 243.15$ K															
0.124[a]	770.4[b]	2.201	785.2	4.453	800.7	7.149	818.2	10.045	836.0	14.612	862.6	19.137	887.4	24.718	915.9
0.131	770.5	2.893	790.0	5.247	805.8	8.305	825.4	11.288	843.5	16.209	871.5	20.440	894.2	27.342	928.7
0.590	773.8	3.970	797.2	5.885	810.1	9.034	829.9	13.302	855.2	17.503	878.6	22.797	906.3	30.052	941.6
1.519	780.5														
$T = 263.15$ K															
0.303[a]	675.4[b]	2.318	692.2	4.708	711.6	7.152	730.2	10.190	751.9	14.682	781.8	20.843	819.1	27.663	857.0
0.978	680.5	3.120	698.9	4.853	712.8	8.204	737.9	11.537	761.1	16.384	792.5	23.102	832.7	30.160	869.9
1.595	686.1	3.464	701.7	5.994	721.5	9.521	747.3	13.175	772.0	19.206	809.7	25.688	846.3		
$T = 283.15$ K															
0.544[a]	580.5[b]	2.486	600.4	5.224	627.5	7.905	651.6	11.68	682.4	16.127	715.3	20.596	745.5	27.228	786.2
0.700	581.1	3.463	610.3	6.116	635.9	9.035	661.2	13.068	693.0	17.891	727.5	22.995	760.7	30.171	802.9
1.440	589.4	4.278	618.3	7.115	644.7	9.763	667.2	14.382	702.8	19.460	738.1	24.859	772.1		
$T = 298.15$ K															
0.886[a]	508.7[b]	2.507	529.4	4.832	556.8	8.142	592.1	11.702	624.7	16.039	660.0	19.588	686.4	24.987	723.1
1.165	512.1	3.244	538.4	5.991	570.3	8.983	600.1	13.473	639.6	17.544	671.5	20.540	693.1	28.133	742.9
1.589	517.7	3.845	545.6	7.217	582.9	10.088	610.3	15.303	654.3	17.618	672.0	22.556	707.0	29.909	753.5
2.304	526.9	4.467	552.7												
$T = 313.15$ K															
1.331[a]	437.8[b]	4.325	480.9	7.083	516.5	10.104	550.0	13.976	587.8	18.704	627.9	23.251	662.5	26.989	688.5
3.246	465.4	4.844	488.3	8.110	528.4	12.272	571.6	15.978	605.5	20.200	639.7	25.257	676.6	29.218	703.1
3.794	473.4	5.970	501.9	9.289	541.4	13.532	583.5	17.421	617.5						
$T = 333.15$ K															
2.141[a]	343.5[b]	5.967	412.2	9.292	461.0	13.489	510.9	16.197	538.7	19.141	566.0	22.689	596.2	27.535	633.2
4.329	382.0	6.850	427.0	10.267	473.5	14.661	523.2	17.857	554.4	20.733	579.8	24.987	614.2	30.141	651.6
4.950	394.5	7.985	443.0	12.031	494.7										
$x = 0.8447$															
$T = 243.15$ K															
0.101[a]	848.5[b]	4.610	878.2	7.333	894.9	8.895	904.2	11.377	918.7	15.678	942.6	21.064	970.7	25.879	994.5
0.608	847.8	4.860	879.7	7.754	897.4	9.610	908.5	12.970	927.8	17.428	952.1	22.893	979.8	27.520	1002.3
1.200	855.8	5.295	882.4	8.051	899.2	10.318	912.6	14.873	938.2	18.512	957.7	25.259	991.5	29.588	1011.8
3.903	873.5	5.787	885.5												
$T = 263.15$ K															
0.246[a]	752.7[b]	2.043	766.6	3.477	777.7	6.885	802.9	10.032	824.5	14.397	852.7	18.878	879.4	24.671	911.8
0.789	756.5	2.121	767.1	4.779	787.6	7.714	808.7	11.532	834.4	16.107	863.1	20.631	889.5	27.284	925.6
1.224	760.0	2.670	771.4	5.814	795.2	9.092	818.2	12.798	842.6	17.489	871.4	22.443	899.7	28.519	932.0
$T = 283.15$ K															
0.456[a]	653.5[b]	2.050	668.6	4.269	689.8	6.969	713.3	9.891	737.0	14.722	773.0	20.575	812.3	25.556	842.9
0.669	654.8	2.779	675.7	5.280	698.8	7.885	720.9	11.763	751.4	16.184	783.3	21.292	816.9	27.251	852.8
1.427	662.5	3.467	682.1	6.065	705.6	8.938	729.5	13.245	762.4	18.997	802.2	22.098	822.0	29.452	865.3
$T = 298.15$ K															
0.721[a]	579.6[b]	2.198	597.2	4.223	620.1	7.884	656.4	13.029	701.2	17.482	735.6	22.337	769.6	27.617	803.6
0.739	579.8	2.768	603.7	6.218	640.4	9.990	675.5	14.581	713.6	19.039	746.9	24.801	785.9	29.954	817.7
1.564	589.7	3.564	612.6	7.197	649.9	11.473	688.4	15.914	723.9	20.402	756.4				

Table 1 (Continued)

p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{-1})	p/MPa	u/(m·s^{1})	p/MPa	u/(m·s^{-1})	p/MPa p	u/(m·s^{-1})
							T = 313.15 K								
1.083[a]	505.8[b]	2.814	530.1	4.857	557.0	8.090	594.1	11.429	627.8	15.873	667.5	20.526	704.5	27.283	752.1
1.615	512.8	3.704	542.1	5.886	569.4	9.296	606.7	13.561	646.4	17.446	680.4	22.223	717.0	30.365	772.1
1.939	517.6	4.213	549.0	6.971	581.9	10.105	614.9	14.550	656.2	18.923	692.2	24.954	736.3		
							T = 333.15 K								
1.750[a]	405.7[b]	5.795	474.2	9.076	520.7	13.341	569.5	16.194	598.1	19.315	626.6	22.643	654.8	27.523	691.4
3.684	439.8	7.461	497.6	10.006	532.2	14.550	581.9	17.594	611.1	20.475	636.5	24.834	671.5	29.932	708.3
4.592	455.6	8.046	505.6	11.555	550.2										

[a] Experimental bubble point pressure.[8] [b] Speed of sound in the saturation liquid was estimated from a polynomial extrapolation at each temperature.

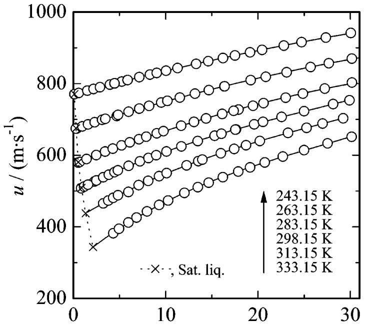

Figure 1. Dependence of the speed of sound u on pressure p in the liquid mixture $0.3555CHF_2CF_3 + 0.6445CHF_2CH_3$.

Table 2. Values of Coefficients of Equation 1

a_i, b_j	$x = 0.3806$	0.6445	0.8447
a_0	1.80023×10^{3}	1.88315×10^{3}	1.98165×10^{3}
a_1	-8.38250	-8.57957	-8.96161
a_2	9.49379×10^{-3}	9.54188×10^{-3}	9.92497×10^{-3}
a_3	-2.18073×10^{-3}	-2.18215×10^{-3}	-2.22444×10^{-3}
b_1	8.12613	7.39177	6.71278
b_2	-0.140038	-0.129287	-0.124789
b_3	7.06919×10^{-4}	6.72389×10^{-4}	7.69771×10^{-4}
b_4	6.45740×10^{-3}	5.12868×10^{-3}	4.22085×10^{-3}
b_5	-1.86650×10^{-4}	-1.55500×10^{-4}	-1.39020×10^{-4}
b_6	1.35378×10^{-6}	1.16486×10^{-6}	1.16784×10^{-6}
δ_{mean}[a]	0.0790	0.1114	0.0825
n	126	131	132

[a] $\delta_{\text{mean}} = 100\Sigma|(u_{\text{exptl}} - u_{\text{calcd}})/u_{\text{calcd}}|/n$; n = number of data points.

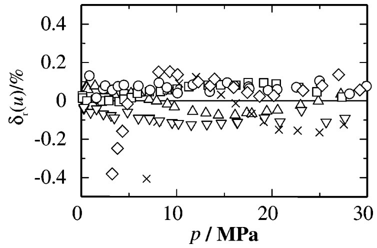

Figure 2. Deviations $\delta_r(u) = 100\ (u_{\text{exptl}} - u_{\text{calcd}})/u_{\text{calcd}}$ of experimental values of the speed of sound u_{exp} from eq 1 (u_{calcd}) for the liquid mixture $0.3555CHF_2CF_3 + 0.6445CHF_2CH_3$. △, 243.15 K; ▽, 263.15 K; □, 283.15 K; ○, 298.15 K; ◇, 313.15 K; ×, 333.15 K.

the largest values of the derivative $(\partial u/\partial p)_T)$ is observed in the vicinity of the liquid−vapor coexistence curve. Similar temperature and pressure dependences of the speed of sound are observed for other compositions. The measured values of the speed of sound are represented for each composition as a function of temperature T/K and pressure p/MPa

$$u_{(T,p)} = \frac{\sum_{i=0}^{2} a_i T^i \sum_{j=1}^{3} b_j p^j}{A + \sum_{j=4}^{6} b_j p^{(j-3)}} \tag{1}$$

where $u_{(T,p)}$/(m·s^{-1}) is the speed of sound at temperature T and pressure p and $A = 1 + a_3 T$. The values of coefficients a_i and b_j were estimated for each measured mixture by a least-squares analysis of all experimental points weighted equally and are listed in Table 2.

Figure 2 is the plot of deviations of experimental values of the speed of sound obtained for the mixture with x = 0.6445 from eq 1. As can be seen in this Figure, the equation reproduces the experimental speed of sound values to within ±0.2% in the high-density region. Rather larger deviations are observed in the low-pressure region for T = 313.15 K and T = 333.15 K. Similar plots were obtained for two other mixtures where the deviations of a few data points exceed ±0.2%. (Maximum deviations are within ±0.4%.) The sound wave generated in a fluid for speed measurements is absorbed significantly in the region close to the critical point by the thermal motion of a molecule. The electric power used in the apparatus was adjusted to the optimum condition (low power) to measure the speed of sound under adiabatic conditions. Therefore, speed of sound values lower than 400 m·s^{-1} where a large absorption appeared for the mixtures investigated were rejected from the fitted data sets. The speed of sound values for the saturated liquid (indicated in Table 1) were estimated for each isotherm by an extrapolation to the bubble-point pressure[8] of data obtained in a narrow pressure interval close to the coexistence region.

Experimental studies on the speed of sound for the binary liquid mixture of hydrofluorocarbons investigated are scarce. Estimates provided by the REFPROP program[9] might be a source of information. This program gives, for example, a value of 511.3 m·s^{-1} at 298.15 K in the saturated liquid (1−0.6445) CHF_2CF_3 +0.6445 CHF_2CH_3, which differs from the present value (508.7 m·s^{-1}) by 2.6 m·s^{-1}. The observed values at elevated pressures are consistent with the estimated values; for example, the differences are −0.3 m·s^{-1} and −1.1 m·s^{-1} at 10 MPa and 20 MPa, respectively. In our previous work,[10] we checked the performance of our instrument by measuring the speed of sound in liquid tetrachloromethane at temperatures from 283.15 K to 333.15 K and pressures up to 30 MPa and confirmed the uncertainty in the experimental results to within ±0.2% by comparing with the selected reference data. Therefore, we assume that the uncertainty in the present results is within ±0.2%, at least in the high-density region. The uncertainty of values related to the saturated liquid might be higher because of the extrapolation process.

The dependences of the speed of sound on pressure and composition in binary mixtures of $(1 - x)CHF_2CF_3$ + $xCHF_2CH_3$ and pure components at 298.15 K are shown

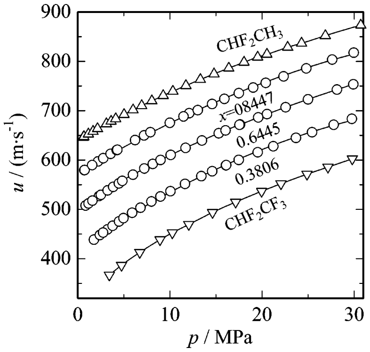

Figure 3. Dependence of the speed of sound u on pressure p at 298.15 K in the liquid system $(1-x)CHF_2CF_3 + xCHF_2CH_3$. ▽, $x = 0$; ○, $x = 0.3806, 0.6445, 0.8447$; △, $x = 1$.

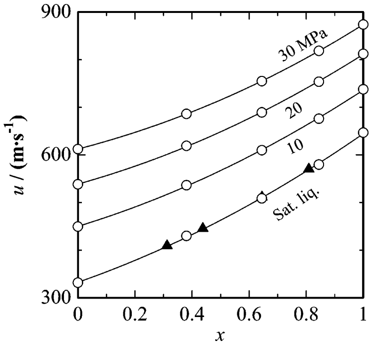

Figure 4. Dependence of the speed of sound u on mole fraction x in the liquid system $(1-x)CHF_2CF_3 + xCHF_2CH_3$ at 298.15 K. ○, This work; ▲, REFPROP.[9]

in Figures 3−5. As can be seen in the Figures, the values of the speed of sound increase with increasing mole fraction. Critical temperatures T_c and pressures p_c of the pure components are 339.33 K and 3.629 MPa (CHF_2CF_3) and 386.41 K and 4.517 MPa (CHF_2CH_3). Because the reduced temperature $T_r = T/T_c$ of CHF_2CF_3 is close to unity in the experimental temperature range, the speed of sound in pure CHF_2CF_3 and the mixture rich in CHF_2CF_3 depends more on pressure, especially in the region close to the coexistence line. The speed of sound increases with increasing mole fraction x, as shown in Figure 4. The shape of $u(x)$ curves is convex (i.e., the apparent excess speed of sound $\Delta u = u_{mix} - \{(1-x)u_1 + xu_2\}$ is a negative and slightly dependent on pressure.

Figure 5 compares the composition dependence of the speed of sound in a saturated liquid for the present system with those for $CHF_2CF_3 + CF_3CH_2F$ [5] and $CHF_2CF_3 + CH_3CF_3$ [6] at 298.15 K reported in our previous works. The composition dependences for $CHF_2CF_3 + CF_3CH_2F$ and $CHF_2CF_3 + CH_3CF_3$ mixtures are nearly straight lines (Δu is close to zero), whereas that for $CHF_2CF_3 + CHF_2CH_3$ is a convex curve over the whole composition range. Despite the fact that the shapes of the composition dependences are different, the dependence of Δu on pressure is similarly moderate for all hydrofluorocarbon systems. However, in the case of systems of a nonpolar component with either a nonpolar or weakly polar component such as hydrocarbon mixtures (for example, benzene + cyclohexane[11] and benzene + isomeric xylenes,[12] reported in the previous papers), the Δu values are more dependent on pressure. It is therefore likely that the apparent excess speed of sound Δu for the mixtures of highly polar hydrofluorocarbons presented here is moderately dependent on pressure because of strong polar−polar intermolecular interactions.

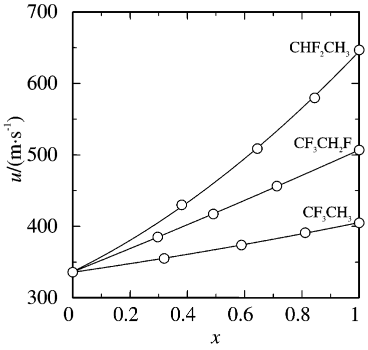

Figure 5. Comparison of the speed of sound u in saturated liquids at 298.15 K for $(1-x)CHF_2CF_3 + xCHF_2CH_3$, $(1-x)CHF_2CF_3 + xCF_3CH_2F$,[5] and $(1-x)CHF_2CF_3 + xCF_3CH_3$.[6]

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