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Ultrasonic Speeds in Compressed Liquid and Vapor Pressures for 1,1,1,2-Tetrafluoroethane

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Ultrasonic speeds in the liquid phase of 1,1,1,2-tetrafluoroethane (CF_3CH_2F) have been measured from 243.11 K to 333.15 K and from near the saturation line to about 30 MPa. The measurements were made using a sing-around technique employing a fixed path acoustic interferometer operated at a frequency of 2 MHz. The probable uncertainty in the results was no greater than ±0.2% except in the low-density region at near the saturation line at higher temperatures. The vapor pressures have also been observed to within ±10 kPa by monitoring the acoustic signal at vapor−liquid equilibrium. When these results were combined, the ultrasonic speeds for the saturated liquid were estimated to within ±1 $m\cdot s^{-1}$.

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1. Introduction

Hydrofluorocarbon, 1,1,1,2-tetrafluoroethane (CF_3CH_2F, HFC-134a), is rapidly replacing dichlorodifluoromethane (CCl_2F_2, CFC-12) in engineering applications and has recently been used as the refrigerant for automotive air conditioning and refrigeration. Therefore, many papers on the various thermophysical properties for CF_3CH_2F have previously been reported (McLinden, 1990; *Thermodynamic Table*, 1990; Penoncello *et al.*, 1995).

The ultrasonic speeds in the liquid phase of CF_3CH_2F were obtained by direct measurement or by estimation from the equation of state (Guedes and Zollweg, 1992; Vstushaninc and Grebenkov, 1993).

In the case of refrigerants, the thermophysical properties near the saturation line are extremely important for process design. In this paper, the apparatus used in the earlier work (Takagi, 1993) was partially modified, and the ultrasonic speed in compressed liquid and vapor for 1,1,1,2-tetrafluoroethane (CF_3CH_2F, HFC-134a) was measured along eight isotherms from 243 K to 333 K at near the saturation line to about 30 MPa. These results were discussed together with those reported previously elsewhere.

2. Experimental Section

Methods. The method used for measuring the ultrasonic speed, u, was a sing-around technique operated at a frequency of 2 MHz similar to that used in previous work (Takagi, 1993). Since the acoustic waves excited in the fluid are significantly absorbed, especially for refrigerants near the saturation line, an interferometer was modified, in part, to improve the signal/noise ratio, as shown in Figure 1.

The diameter of the transducer holder fixed in parallel with the reflector was extended to twice the transducer diameter to avoid the influence of noise caused by the reflection from the side wall. Moreover, the dead volume in the holder above the transducer was reduced.

A piston cylinder for oil−sample separation was employed in a pressure vessel made of SUS306 stainless steel. The volume change in the sample chamber when the piston was moved full range was from 40 to 60 cm^3. The pressure seal used for the interferometer and piston cylinder was a silicon-containing rubber O-ring for low temperature, blended specially by Mitsubishi Cable Industrials Ltd. The electrode for signal transmission was separated from the thermostat to avoid the influence caused by electrical leakage due to the formation of frost at low temperature. The acoustic interferometer, A in Figure 2, was immersed in a liquid thermostat containing a mixture of ethylene glycol and water; 45/55 vol %, controlled to within ±20 mK.

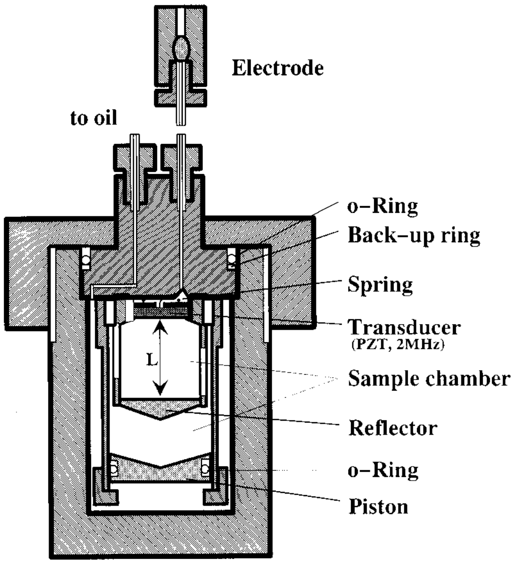

Figure 1. Acoustic interferometer.

The temperature was observed by a quartz thermometer, which was calibrated with a standard platinum thermometer to within ±5 mK against ITS-90. The pressure generated by a hand oil pump using a silicon oil of 1×10^{-4} $m\cdot s^{-1}$ in viscosity was transmitted to the sample through a piston moving in the interferometer, A, and/or piston cylinder, B. The pressure in the sample was observed through the strain of the capsule, C in Figure 2, by two precision strain gauges of maximum pressure: (5 ± 0.003) MPa (Nagano Keiki Ltd. KH-17) and (35 ± 0.005) MPa (Tsukasa Sokken Co. HP-22-G) calibrated by a dead weight tester through a digital voltmeter, DVM (Advantest Ltd., R6551). Figure 3 is a sample−oil separator employing a Teflon capsule of 0.2 mm thickness. The flexible chamber was packed with silicon oil as the pressure transmitting medium, which was held at about 353 K to minimize the

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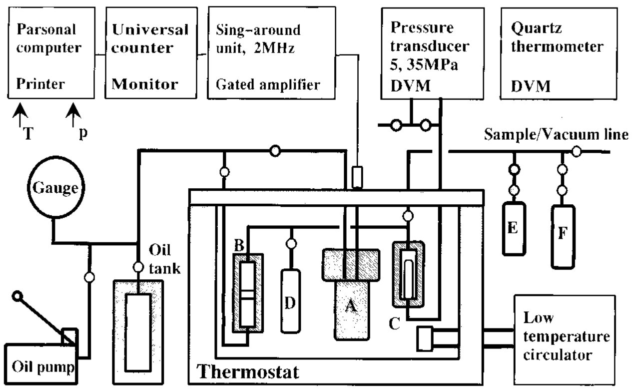

Figure 2. Schematic diagram of the experimental apparatus: A, acoustic interferometer; B, piston cylinder; C, sample–oil separator; D, E, F, sample cylinders.

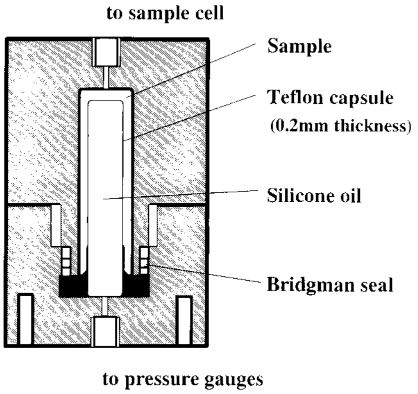

Figure 3. Sample–oil separator employing a Teflon capsule used for pressure measurement.

effect of thermal expansion. The sensitivity in the capsule strain was equivalent to an accuracy of within ±0.001 MPa in pressure.

The ultrasonic speed, u, 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, between the transducer and reflector using the following equation.

$$u = 2L/(T_2 - T_1) \tag{1}$$

The period, T, was observed by a universal counter (Advantest Ltd., TR-5822) with resolution of 0.1 ns as the average value of 1000 periods. The value of $2L = (47.602 \pm 0.004)$ mm was determined by measuring the period, T, in pure tetrachloromethane, CCl_4, at the low u value in organic liquid: 921.11 $\mathrm{m{\cdot}s^{-1}}$ at 298.15 K and 0.1 MPa, reported by Tamura *et al.* (1983). The influence of L due to the temperature changes was calculated from the expansivity, α $(=13.6 \times 10^{-6}\ \mathrm{K^{-1}})$ of SUS306. That for the compressibility of stainless steel was omitted because it can be neglected in the range of pressures up to 30 MPa.

The acoustic wave in the fluid excited for u measurement was absorbed much more readily in the gas phase than in the liquid phase. Using this phenomenon, the vapor pressure was estimated by monitoring the acoustic signal on an oscilloscope, that is, when the liquid level at vapor–liquid equilibria was located to the bottom position of the transducer.

The temperature of the strain gauge transducer was maintained in the thermostat by a water jacket. The pressure gauge with a measuring range of 5 MPa has a resolution of 800.00 mV/MPa with a stability of ±0.1 mV corresponding to ±0.12 kPa.

Materials. A high-quality sample of CF_3CH_2F was supplied by Daikin Industrials Ltd. Their purity was better than 99.96+ mol %, as measured by glc.

3. Results and Discussion

The experimental ultrasonic speeds, u, in the liquid phase of 1,1,1,2-tetrafluoroethane (CF_3CH_2F) are listed in Table 1 and illustrated graphically in Figure 4.

In this work, the measurements near the saturation line have been carried out in detail at small pressure intervals along eight isotherms. The results increase monotonously with increasing pressure, and the pressure effect, $(\partial u/\partial p)_T$ becomes greater with increasing temperature, especially in the saturation region, as shown in Figure 4. The critical temperature, T_c, and pressure, p_c, for CF_3CH_2F are 374.27 K and 4.0623 MPa, respectively (Sato *et al.*, 1994). It is well-known that close to the critical point, the absorption of the acoustic wave too rapidly increases due to the thermal motion in the molecules. This effect approaches maximum at the critical point, and the speed approches a minimum around 100 $\mathrm{m{\cdot}s^{-1}}$ (Bhatia, 1967). For the present compound, the large absorption was also observed at 333 K, near the critical point, especially in the saturation region for the u data below 400 $\mathrm{m{\cdot}s^{-1}}$. This phenomenon was pronounced in the second echo, and therefore these results included a larger error than those in the high-density region. No data under higher pressure of (243 and 253) K are given in Table 1. The pressure could not be transmitted to the sample above these conditions because the piston would not move in the cylinder. It appears that the O-ring used as the seal in the piston swelled or hardened with decreasing temperature.

The u data at each temperature are well represented as a smoothed function of temperature, T, and pressure, p, by the following polynomial equation:

$$u/\mathrm{m{\cdot}s^{-1}} = \sum_{i=0}^{4}\sum_{j=0}^{4} a_{ij}[T/\mathrm{K} - 273.15]^i[p/\mathrm{MPa} - 5]^j \tag{2}$$

The values of the coefficients, a_{ij}, were calculated by a weighted least-squares analysis of data listed in Table 2 with a mean error, δ_{mean}. In this determination, the u values lower than 400 $\mathrm{m{\cdot}s^{-1}}$ exhibited large acoustic absorption and those at 333.108 K were removed from the data set used in the least-squares fit. The experimental values are in good agreement with the u estimated from eq 2 with deviations not exceeding ±0.2%. The largest deviations are in the vicinity of the saturation line at high temperature, where the value is particularly sensitive to pressure changes.

For CF_3CH_2F, ultrasonic speeds in the liquid phase have been measured by Guedes and Zollweg (1992) in the ranges from 180 K to 380 K and pressures up to 70 MPa by using a pulse echo overlap method. Vstushaninc and Grebenkov (1993) also measured the u values from 220 K to 320 K and from 0.1 MPa to 20 MPa by a pulse method employing double transducers. The results, u, of this work were compared with their data at 243.15 K and 298.15 K in Table 3.

At 243.15 K, the ultrasonic speeds of the present work are slightly lower by about 2–3 $\mathrm{m{\cdot}s^{-1}}$ than those reported by Guedes and Zollweg (1992) and Vstushaninc and Grebenkov (1993) for low pressure, but the values of this work under 10 MPa are in good agreement with their experimental values. The three data sets at 298.15 K are in agreement with each other but exhibit a slightly different pressure effect, $(\partial u/\partial p)_T$. To check the reproducibility

Table 1. Ultrasonic Speeds, *u*, in the Liquid Phase for 1,1,1,2-Tetrafluoroethane at Various Pressures

p/MPa	u/m·s^{-1}	p/MPa	u/m·s^{-1}	p/MPa	u/m·s^{-1}	p/MPa	u/m·s^{-1}
T = 243.151 K							
0.182	761.8	1.135	767.6	5.068	790.8	9.939	817.5
0.336	762.7	1.514	770.0	6.261	797.5		
0.526	763.9	2.046	773.1	7.184	802.7		
0.683	764.9	3.184	779.9	8.028	807.3		
0.825	766.1	3.512	781.9	9.033	812.7		
T = 253.147 K							
0.130	714.6	1.395	723.2	4.407	742.5	10.006	776.0
0.279	715.7	1.854	726.2	5.277	747.8	11.360	783.6
0.434	716.6	2.427	729.9	6.210	753.6	12.864	791.9
0.720	718.8	3.316	734.9	7.253	759.9	13.665	796.3
0.854	719.7	3.892	739.3	8.227	765.7		
T = 263.142 K							
0.197	668.4	2.411	684.8	9.127	729.2	19.638	789.3
0.489	670.5	2.935	688.5	10.087	735.3	20.592	794.2
0.628	671.8	3.589	693.1	11.341	742.8	24.050	811.7
0.826	673.0	4.227	697.5	12.940	752.2	25.153	817.2
0.859	673.5	5.135	703.6	14.389	760.6	26.615	824.3
1.038	674.8	6.073	709.8	15.913	769.2	27.794	829.9
1.356	677.2	6.878	715.1	17.554	778.2	30.163	841.0
1.792	680.4	8.216	723.6	18.239	781.8		
T = 273.137 K							
0.299	622.5	3.220	646.3	8.900	686.8	17.111	738.0
0.509	624.2	3.519	648.6	10.101	694.8	17.987	743.0
0.769	626.7	4.109	652.9	10.588	698.0	19.295	750.3
0.876	627.6	4.647	657.1	12.212	708.4	20.495	757.0
1.151	629.9	5.140	660.7	13.064	713.6	22.573	768.3
1.473	632.7	5.863	665.8	13.947	719.2	24.962	780.9
2.107	637.8	6.863	672.9	14.953	725.3	27.083	791.7
2.554	641.4	7.899	680.0	16.200	732.7	29.681	804.5
T = 283.133 K							
0.413	576.5	3.082	600.5	8.214	641.1	19.118	713.3
0.589	578.2	3.569	604.6	9.206	648.4	20.994	724.2
0.710	579.3	4.019	608.4	10.963	660.6	22.792	734.4
1.087	582.8	4.363	611.3	11.447	664.2	24.820	745.6
1.454	586.2	5.186	617.7	12.906	674.1	26.978	757.2
1.980	590.9	6.106	625.0	14.306	683.3	29.696	771.4
2.388	594.6	7.044	632.3	15.987	694.1		
2.469	595.2	7.061	632.4	17.319	702.4		
T = 283.135 K[a]							
0.418	576.9	4.927	616.1	15.187	689.4	24.875	747.8
0.792	580.3	7.488	635.8	17.588	704.3	27.141	758.2
1.338	585.4	10.182	655.6	20.474	721.2	30.126	773.5
2.999	600.0	12.654	672.6	22.400	732.4		
T = 298.125 K							
0.672	507.6	4.727	549.6	13.132	619.7	24.818	696.3
0.834	509.8	5.431	556.2	14.482	629.5	26.988	708.8
1.006	511.8	6.219	563.0	15.877	639.4	29.567	722.1
1.597	518.2	7.249	572.5	16.795	645.7		
1.976	522.3	8.125	580.0	17.332	649.4		
2.456	527.2	9.150	588.6	18.747	658.7		
3.160	534.4	10.110	596.4	19.946	666.5		
3.910	541.6	11.639	608.4	22.620	682.6		
T = 313.118 K							
1.018	437.8	5.042	488.1	11.056	547.8	19.685	615.2
1.382	443.1	5.320	491.2	11.754	553.8	21.905	630.4
1.486	444.5	5.883	497.4	12.903	563.5	24.234	645.7
2.065	452.2	6.500	504.1	13.836	571.2	26.936	662.5
2.496	457.8	7.091	510.4	14.839	579.2	29.728	678.6
3.128	465.8	7.821	517.5	15.824	586.8		
4.13	477.8	8.200	521.2	17.006	595.9		
4.478	481.8	8.868	527.6	17.741	601.3		
T = 333.108 K							
1.679	341.2	5.569	407.6	12.99	494.2	20.773	560.5
2.083	349.7	6.036	414.7	13.984	503.7	22.934	576.8
2.738	362.4	7.313	431.6	15.001	513.0	23.731	582.5
3.083	368.8	8.019	440.5	15.839	520.5	25.720	596.4
3.595	377.1	8.995	452.0	16.915	529.8	28.008	611.7
3.901	382.9	9.791	461.0	17.219	532.2	30.673	628.8
4.332	389.9	10.921	473.2	17.982	538.6		
4.981	399.5	11.965	484.0	18.918	546.3		

[a] Values remeasured with another sample.

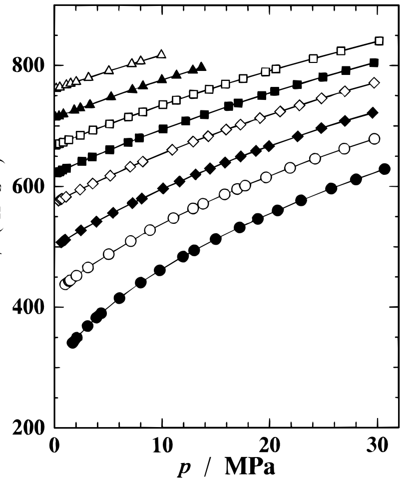

Figure 4. Pressure dependence of ultrasonic speeds in the liquid phase of 1,1,1,2-tetrafluoroethane: (△) 243.15 K; (▲) 253.15 K; (□) 263.14 K; (■) 273.14 K; (◊) 283.13 K; (◆) 298.12 K; (○) 313.12 K; (●) 333.11 K.

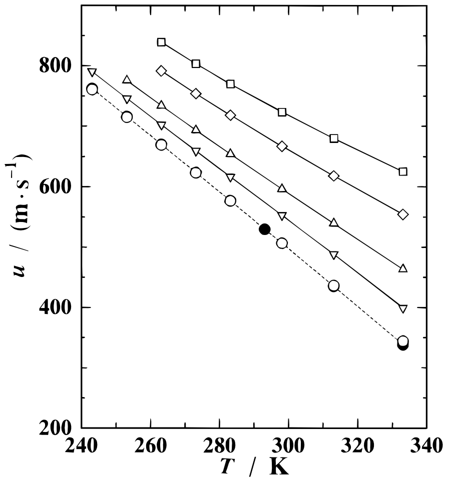

Figure 5. Temperature dependence of ultrasonic speed in the liquid phase of 1,1,1,2-tetrafluoroethane. Saturation line: (○) this work; (- - -) calculated from eq 4; (●) Guedes and Zollweg (1992). Calculated from eq 2: (▽) 5 MPa; (△) 10 MPa; (◊) 20 MPa; (□) 30 MPa.

Table 2. Coefficients, a_{ij}, for Eq 2 and Mean Error, δ_{mean}, of Calculated Ultrasonic Speeds from Experimental Values in Liquid 1,1,1,2-Tetrafluoroethane[a]

	i			
j	0	1	2	3
0	6.594304E 2	−4.311815E 0	1.024169E-3	−1.754486E-5
1	7.313431E 0	7.052815E-2	6.792333E-4	
2	−9.386462E-2	−2.825638E-3	−2.263787E-5	
3	2.399543E-3	9.490643E-5	−1.622986E-6	
4	−3.990830E-5	−1.453682E-6	1.558093E-9	

[a] $\delta_{mean} = 0.032$, $\delta_{mean} = |100[(u_{exp} - u_{calc})/u_{calc}]|/n$, where n (=207) is the number of data points.

of experimental values, the speed in the new sample was also measured at 283.135 K. At all pressure conditions, the values of u were reproduced within ±0.13% with those at 283.133 K, as shown in Table 1. From these facts, the uncertainty in the present u data in the range 243 K to 333 K was estimated to be within ±0.2% except at values lower than 450 m·s^{-1}. Piao *et al.* (1990) have developed a modified Benedict−Webb−Rubin equation of state for CF_3CH_2F, and the ultrasonic speeds derived from the equation were presented in the databook published by the Japan Association of Refrigeration (1990). These results

Table 3. Comparison of Ultrasonic Speeds, u/m·s^{-1}, in Liquid 1,1,1,2-Tetrafluoroethane at 243.15 K and 298.13 K with Literature Values[a]

	T=243.15 K				T=298.15 K			
p/MPa	$u_{(T)}$	$u_{(G)}$	$u_{(V)}$	$u_{(P)}$[b]	$u_{(T)}$	$u_{(G)}$	$u_{(V)}$	$u_{(P)}$
2	771.0	774.1	773.1	717.7	521.4	522.0	523.8	519.7
4	782.9	785.9	784.8	736.3	542.0	542.6	543.1	542.9
6	794.5	797.5	796.1	754.1	560.9	561.5	561.4	564.1
8	805.9	808.7	807.1	771.1	578.7	579.2	578.9	583.7
10	817.0	819.6	817.7	787.6	595.2	595.8	595.4	602.0
15		845.4	842.7	826.2	632.8	633.7	632.5	643.0
20		869.6	865.7		666.0	667.4	663.9	

[a] $u_{(T)}$, this work; $u_{(G)}$, Guedes and Zollweg (1992); $u_{(V)}$, Vstushaninc and Grebenkov (1993); $u_{(P)}$, derived from equation of state (1990). [b] At 245.00 K.

Table 4. Comparison of Vapor Pressure, p_s/MPa, for 1,1,1,2-Tetrafluoroethane with Literature Values[a]

T/K	$p_{s(T)}$	$p_{s(calc)}$	$p_{s(S)}$	$p_{s(B)}$
243.15	0.0832 ± 0.0042	0.0823	0.0844	
253.15	0.128 ± 0.0018	0.130	0.132	
263.14	0.197 ± 0.0024	0.197	0.200	
273.14	0.289 ± 0.0033	0.288	0.293	
283.13	0.410 ± 0.0038	0.410	0.414	
298.13	0.666 ± 0.0052	0.661	0.665	0.663
313.12	1.009 ± 0.0046	1.012	1.015	1.013
333.11	1.673 ± 0.0055	1.678	1.679	1.676

[a] $p_{s(T)}$, this work; $p_{s(calc)}$, calculated from eq 3; $p_{s(S)}$, Sato *et al.* (1994); $p_{s(B)}$, Baehr and Tillner-Roth (1991).

Table 5. Comparison of Ultrasonic Speed at Saturation Line, u_s/m·s^{-1} for 1,1,1,2-Tetrafluoroethane with Literature Values[a]

T/K	$u_{s(T)}$	$u_{s(calc)}$	$u_{s(G)}$	$u_{s(V)}$	$u_{s(P)}$
243.15	759.5	759.6	762.6	758.9	643.7
253.15	714.4	714.3	716.2	713.1	674.9
263.14	668.8	668.6	669.9	667.4	641.3
273.14	622.7	622.7	623.5	621.8	604.2
283.13	576.3	576.5	576.9	576.4	563.9
298.13	506.8	506.7	506.5	509.1	502.9
313.12	436.4	436.4[b]	435.2		437.0
333.11	341.0	342.1[b]	338.3		341.3

[a] $u_{s(T)}$, this work; $u_{s(calc)}$, calculated from eq 4; $u_{s(G)}$, Guedes and Zollweg (1992); $u_{s(V)}$, Vstushaninc and Grebenkov (1993); $u_{s(P)}$, calculated from equation of state (1990). [b] Extrapolated values.

indicate different absolute values and $(\partial u/\partial p)_T$ from the experimental values. This may be caused by difficulties in the estimation of ultrasonic speed from the wide-range equation of state covering the gas, liquid, and supercritical regions.

In order to determine the ultrasonic speed for the saturated liquid, the vapor pressure, p_s, was measured by monitoring the liquid level. That is, when the liquid level in the liquid−vapor equilibria apparatus was at the bottom position of the transducer, the pressure was measured at the condition by asssuming the fluid was entirely liquid, with a very small amount of gas present as a source of error. The mean results of vapor pressure for CF_3CH_2F in four or five data points and the differences from each value are listed in Table 4. There are many souces of accurate vapor pressure values for this compound. Sato *et al.* (1994) reported the vapor pressure $p_{s(S)}$, which was evaluated from many data sets, as most reliable values. In Table 4, our extensive experimental results are in excellent agreement with the $p_{s(S)}$ and with the $p_{s(B)}$ values of Baehr and Tillner-Roth (1991), which were measured with an accuracy of ±0.1 kPa.

The vapor pressure results were correlated in the temperature range from 243 K to 333 K using the following modified Wagner equation:

$$\ln(p_r) = 1(T_r)[-7.5228(1 - T_r) + 1.23489(1 - T_r)^{1.5} - 3.57713(1 - T_r)^3] \quad (3)$$

where p_r ($=p_s/p_c$) is the reduced pressure, $p_c = 4.0623$ MPa, and T_r ($=T/T_c$) is the reduced temperature, $T_c = 374.27$ K. This equation reproduced the experimental values within ±10 kPa. When the coefficients in Table 2 and the vapor pressures in Table 4 were combined, the ultrasonic speeds, u_s, for the saturated liquid were estimated in the range from 243 K to 298 K by extrapolation to the vapor pressure, p_s. These values are given in Table 5 together with the reference values. The resuts were represented by the following equation:

$$u_s/\mathrm{m{\cdot}s^{-1}} = 2088.98 - 75.4732T_r^{-1} - 1867.45T_r \quad (4)$$

where $T_r = T/T_c$. The values calculated by the above equation agree with the extrapolated values within ±1 m·s^{-1}. As described above, the measurements near the saturation line at high temperatures were accompained by a large irregularity in the ultrasonic speeds based on a strong absorption of the acoustic wave. Therefore the u_s values at 313 K and 333 K were estimated by using those at pressures lower than 10 MPa. These u_s values are also in good agreement with those calculated from an equation given by Guedes and Zollweg (1992) and those measured by Vstushaninc and Grebenkov (1993). The results derived from the equation of state are in an agreement near room temperature but at low temperature are lower than these experimental results. In many cases the equation of state covers wide ranges of temperatures and pressures including the critical region, and moreover the estimation of ultrasonic speed from the equation of state is complicated. It seems reasonable that these errors in the calculated values may be responsible for the differences in u between experimetal and estimated values. Accurate ultrasonic speeds determined experimentally are most of those used to investigate the reliability of the equation of state.

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