1120	Water		· · · ·
k	Boltzmann's constant	exp	exp
m	number of components in the mixture	ร่	satu
MEA	ethanolamine	spt	valu
p	pressure	ω	valu
R	universal gas constant ($R = 8.3143 \text{ J mol}^{-1} \text{ K}^{-1}$)	,	liqui
Τ	temperature	"	Van
V	molar volume	•	rofo
V	volume	• /	solu
x	mole fraction in liquid phase	~	Solu
v	mole fraction in vapor phase	θ	star
,		Req	gistry No.
Greek	Symbols		
α,	polarizability of component i	Liters	ature Cite
\mathbf{v}_{i}	activity coefficient for component i dissolved in		
113	component /	(1)	Kling, G.; M
4	Lennard-Jones energy parameter	(2)	Kling, G. Ph
n. n.	reduced densities	(3)	1988.
יויי יו <i>וי</i> יוי יווי		(4)	Nath, A.; Be
'/2' '/	3 chemical potential, dipole moment	(5)	Bender, E.
μ	moler depet	(6)	1960 32
p 	molar density	(7)	Lebowitz, J
ρ.	number density	(0)	43, 774.
σ	Lennard-Jones distance parameter	(8)	Shoor, S. K Hirschfeider
φ	fugacity coefficient	(0)	es and Liqu
φ	volume fraction in liquid phase	(10)	Liabastre, A
Subscr	ints	(11)	1974. McCiellan
Cubboon		(11)	man and Co
C	critical value	(12)	Saul, A.; W
ij	binary interactions or attractive property between	(13)	McGarry, J
	molecules of species <i>i</i> and <i>j</i>	(14)	aminlösuno
J	component /		der Technis
H ₂	hydrogen		
mix	mixture	0	
pot.	value resulting from intermolecular forces	Receiv	illy acknowle
v	repulsive property	Forsch	ungsgemeir

Superscripts

HXD	exceri	nenia	VHILLE

irated

- e calculated from the scaled particle theory
- e at infinite dilution
- d phase
- or phase
- erred to number of particles, reduced
- te free
- idard value ($p^{\Theta} = 0.1 \text{ MPa}$)

H₂, 1333-74-0; ethanolamine, 141-43-5.

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Ultrasonic Speed for Liquid Trichlorofluoromethane and 1,1-Dichloro-2,2,2-trifluoroethane at Temperatures from 283 to 373 K and Pressures up to 75 MPa

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The ultrasonic speeds for liquid trichlorofluoromethane (CCl₃F) and 1,1-dichloro-2,2,2-trifluoroethane (C₂HCl₂F₃) were measured from 283 to 373 K and from 0.1 MPa or their saturated vapor pressure to about 75 MPa. The measurements were carried out by a sing-around technique operated at a frequency of 2 MHz with the uncertainty of $\pm 0.2\%$. The temperature and pressure variation of ultrasonic speed and related properties are discussed.

Introduction

Trichlorofluoromethane, CCl₃F (CFC-11), has a high solubility for polymers and is usually used as a solvent in the foam and textile industries. Due to its ability to destroy ozone, its use as a foaming agent and refrigerant is being curtailed. 1,1-Dichloro-2,2,2-trifluoroethane, C2HCl2F3 (HCFC-123), is considered a suitable replacement compound of CCl₃F (1).

Ultrasonic speeds in fluids, which travel with the compression waves, give a powerful clue for elucidating the thermodyanmic properties. In previous papers (2-4), we have been reported the ultrasonic speed of liquid refrigerants at several conditions of temperature and pressure. This paper describes the new experimental results of ultrasonic speeds in the liquid phase of CCI_3F and $C_2HCI_2F_3$. The variation of the ultrasonic speed and related thermodynamic properties for these fluids as a function of temperature and pressure are discussed and compared with related properties of tetrachloromethane, CCl₄.

Experimental Section

Materials. Trichlorofluoromethane, CCl₃F, and 1,1-dichloro-2,2,2-trifluoroethane, C₂HCL₂F₃, were supplied by Daikin Industrials Ltd. Their purities were better than 99.8 wt %, as measured by GLC. Tetrachloromethane, CCI4, was a "spectral grade" reagent supplied by Dojin Chemical Ltd.; its purity, de-

394 H.O

water

Table I. Physical l	Properties for	Each Con	npound
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compound	CCl4	CCl ₃ F	C ₂ HCl ₂ F ₃
molecular wt	153.82	137.37	152.93
melting temp/K	250.20	162.15	166.15
boiling temp/K	349.90	296.90	300.15
critical constants ^a			
$T_{\rm c}/{\rm K}$	556.4	471.20	456.85
p./MPa	4.56	4.41	3.67
$\rho_{\rm c}/(\rm kg\cdot m^{-3})$	558	554	550
dipole moment/(10 ⁻³⁰ C·m)	0	0.45 ^b	0.8°

^aReference 5 for CCl₄ and CCl₃F; reference 1 for $C_2HCl_2F_3$. ^bReference 6. ^cEstimated by Debye equation (6) using dielectric constant at different temperatures (7).

termined by GLC, was better than 99.99 mass %. The physical properties of each substance are listed in Table I.

Apparatus. The method used for measurement of ultrasonic speed was a sing-around technique employing a fixed-path ultrasonic interferometer, similar to that described previously (2, 8). An acoustic interferometer placed in the high-pressure vessel was immersed in an ethylene glycol bath controlled to within ± 0.02 K between 283 and 333 K and to ± 0.03 K between 343 and 373 K. The temperature in the bath was measured by a quartz thermometer with a resolution of ± 0.001 K. To reduce the temperature variation in the sample caused by the pressure change, the difference in temperature between sample chamber and bath was observed by a T-type thermocouple. The pressure was generated by a hand-operated oil pump and was transmitted to the sample through the moving of a piston, sealed by a Teflon-coated O-ring, in a piston cylinder. The pressure was measured by two strain gauges with ranges of 10 and 100 MPa to within ± 0.03 and ± 0.08 MPa, respectively.

The ultrasonic speed, u, was obtained by measuring the repeat period, t, of a short acoustic pulse traveling over the known distance, L, between the transducer and reflector. In this work, a new acoustic interferometer made of stainless steel (SUS304) with L = 25 mm was used. The acoustic path length was determined by using the speed of sound in pure tetrachloromethane at 298.15 K and 0.1 MPa (924.3 m·s⁻¹) reported by Bobic et al. (9). The variation of L resulting from temperature and pressure was calculated from the coefficients of expansion and compression of the metal (10).

As preliminary work, the ultrasonic speed, u, in CCl₄ was measured. These results, as given in Table II, agree to within $\pm 0.2\%$ with those measured by Bobic et al. (9). Lainez et al. (11) measured the ultrasonic speed at 298.15 K. Their cell was calibrated by using the value in CCl₄ of 920.3 m·s⁻¹ at 298.15 K and 0.1 MPa; hence their results will be lower. However, the pressure dependence in u is in good agreement with the value presented here and in ref 9. The estimated uncertainty in the ultrasonic speeds is $\pm 0.2\%$, with greater uncertainty for values at pressure close to the saturated vapor pressure.

Results and Discussion

The experimental results of the ultrasonic speed, u, in the liquid phase of CCl₃F and C₂HCl₂F₃ at pressure, p, are listed



Figure 1. Pressure dependence, *p*, of ultrasonic speed, *u*. CCl₄: **•**, 298.15 K. CCl₃F: **•**, 298.15 K; **•**, Δ (ref 15), 353.15 K. C₂HCl₂F₃: **•**, 298.15 K; **•**, 353.15 K.

in Table III and are shown graphically in Figure 1. The variation in u as a function of temperature and pressure can be represented by the polynomial equation

$$u/(\text{m}\cdot\text{s}^{-1}) = \sum_{j=0}^{2} \sum_{j=0}^{4} a_{ij} (T/\text{K})' (p/\text{MPa})^{j}$$
 (1)

The values of the coefficients a_{ij} , calculated by least-squares analysis of all experimental data, are presented in Table IV. The maximum and standard deviations of the experimental results from this equation are 0.11 and 0.055% for CCl₃F, and 0.17 and 0.065% for C₂HCl₂F₃, respectively.

The ultrasonic speed in the liquid phase, u_{p_s} , at saturation pressure, p_s , was calculated from eq 1. p_s was derived from the equation reported by Downing (12) for CCl₃F and by Weber (13) for C₂HCl₂F₃. The results are presented in Table III and Figure 2. For C₂HCl₂F₃, these values can be represented by the following equation:

$$u_{p_*}/(\text{m}\cdot\text{s}^{-1}) = 693.7 - 3.4580t + 3.6659 \times 10^{-4}t^2 + 1.0649 \times 10^{-5}t^3$$
 (2)

where t = (T/K) - 298.15. This equation reproduced the extrapolated values to within a maximum deviation of 0.02%.

These are no previous measurements of the ultrasonic speeds in $C_2HCl_2F_3$. For CCl_3F , Chavez et al. (14) obtained the values in the liquid phase along the saturation line from 162 to 458 K by a pulse echo overlap method, given by the dotted line in Figure 2. They give 555.5 m·s⁻¹ as the ultrasonic speed at 353.15 K and p_s . This value is a little lower than our result: 557.2 m·s⁻¹. In the vicinity of the saturation pressure, absorption of the acoustic wave generated in the sample was

Table II. Comparison of the Observed Ultrasonic Speed $u/(m \bullet s^{-1})$ in Tetrachloromethane, CCl₄, and Trichlorofluoromethane, CCl₅F^a

				CCl ₄		_		CCI	F
	T/K = 298.15 $T/K = 343.15$ $T/K = 373.$					73.15	T/K = 353.15		
p/MPa	this work	ref 9	ref 11	this work	ref 9	this work	ref 9	this work	ref 15
0.1	924.3	924.3	920.3	786.9	786.3			594.9*	590.2*
20	998.7	998.1	994.8	878.2	877.8	803.4	803.4	692.9	690.5
40	1062.2	1061.7	1058.7	953.1	952.7	886.6	886.9	789.8	787.5
60	1118.2	1116.5	1114.9	1017.0	1017.2	956.4	956.5	867.6	865.3

^a Values marked with an asterisk were measured at 5 MPa.

Table III.	Ultrasonic Speed	, $u/(m \bullet s^{-1})$, in the 1	Liquid Phase of	Trichlorofluoromethan	e, CCl ₂ F, and
1,1-Dichlo	ro-2,2,2-trifluoroet	hane, C ₂ HCl ₂ F ₃ , at V	Various Pressure	es, p/MPaª	•
			and the second secon		

p/MPa	$u/(m \cdot s^{-1})$	p/MPa	$u/(m \cdot s^{-1})$	p/MPa	$u/(m \cdot s^{-1})$	p/MPa	$\frac{u}{(m \cdot s^{-1})}$	p/MPa	$\mu/(m \cdot s^{-1})$	p/MPa	$\mu/(m \cdot s^{-1})$
		• /	<u> </u>	• /	CC	Cl _a F	, 、 ,	F () ()			
					298.	15 K					
0.1056*	743.5*	8.23	786.9	20.32	843.0	35.10	901.9	49.76	953.2	64.83	1000.4
0.132	743.0	15.66	800.4 822 4	25.45	864.0 883.0	40.27	920.7	55.20 60.21	970.8	70.13	1015.9
5.818	774.5	10.00	022.4	30.11	003.0	40.10	931.0	00.31	900.7	14.11	1029.1
					313	15 K					
0.1734*	692.4**	8.44	741.1	20.49	801.7	35.24	864.2	50.19	921.3	65.50	968.8
3.202	710.9	11.14	755.7	25.85	825.3	40.71	885.1	55.23	936.1	69.48	980.9
5.725	725.8	15.99	780.3	30.27	843.8	45.95	904.2	60.59	953.4	73. 98	994.4
					333.	15 K					
0.3111*	627.5**	8.78	682.1	20.35	746.6	35.95	817.4	49.87	872.6	64.49	923.3
3.176	645.3	11.01	695.6	25.32	771.2	40.48	836.7	54.95	890.9	69.78	940.2
6.343	666.6	15.59	721.8	30.63	795.3	45.28	855.4	59.79	907.5	74.41	954.6
					353.	15 K					
0.5192*	557.4**	8.20	619.4	20.89	697.7	35.38	769.6	50.07	830.8	64.94	884.6
4.621	591.7	11.46	641.1	26.01	724.8	40.92	793.8	54.77	848.5	69.91	901.2
6.442	605.7	15.81	668.5	30.55	747.2	44.97	810.7	60.00	867.4	73.86	914.1
					C₂H	Cl_2F_3					
					283.	15 K					
0.0506*	745.7**	7.077	785.2	21.00	852.1	36.06	913.6	50.91	966.6	65.69	1014.5
0.1	746.0	9.22	796.0	20.10	8067	40.00	931.0	00.00 60.76	982.3	75.50	1030.1
4.537	771.6	15.96	829.4	31.70	030.7	40.10	343.1	00.70	555.0	10.00	1044.0
					203	15 K					
0.0756*	711.0**	7,720	756.4	20.36	819.4	35.00	882.0	50.29	938.8	65.30	988.2
0.132	712.1	10.74	722.5	24.85	839.8	40.51	902.9	54.81	954.2	69.00	999.9
4.026	735.5	12.96	784.0	30.03	861.9	45.07	920.2	60.09	971.7	74.01	1015.1
6.816	751.4	15.90	798.6								
					298.	15 K					
0.0913*	693.7**	7.735	740.1	20.55	806.5	36.01	873.5	51.06	930.0	65.06	977.2
0.464	694.9 701 9	10.88	151.1	20.15	852.1	41.00 45.75	093.3 910.8	00.00 60.46	940.9	09.00 74 44	1006.2
4.000	721.0	10.07	102.2	00.77	002.1		010.0	00.40	002.1		100012
				04 / F	303.	.15 K	001.0	FO 01	010 5	00 10	000 7
0.1094	676.4**	8.99	731.3	21.45	796.3	36.26	801.2 979.0	55 37	910.7	70.82	900.7
6 267	715.2	16.20	772.4	20.31	838.8	45.97	898.9	61.17	952.5	75.59	999.1
0.201		20100					•••••	•	• • • • • •		
0 15 40+	C (1 0++	7 796	604.9	00.01	313.	.15 K	997 1	50.90	902 1	64 71	942 7
0.1043*	641.9**	11 01	7137	20.21	7894	40 10	853.5	55 55	912.2	70.22	961.7
3.855	668.2	15.37	738.4	30.53	812.9	44.83	872.3	60.59	929.6	74.64	975.7
5.435	679.1										
					323	.15 K					
0.2123*	607.7**	6.367	652.6	21.04	741.0	35.83	811.3	50.55	871.0	65.63	925.2
0.487	608.6	12.14	691.2	25.63	764.3	40.70	831.6	55.99	891.0	69.76	939.0
3.827	633.9	15.72	712.2	31.09	790.2	45.18	850.2	61.17	909.5	75.26	956 .7
					333	.15 K					
0.2862*	573.6**	8.95	641.0	21.14	715.8	36.31	79 0.8	50.36	849.3	65.63	905.7
0.505	574.8	11.74	660.4	25.63	739.7	40.72	809.9	55.70	869.8	70.39	921.6
4.153	605.3	16.01	686.9	31.29	767.6	45.51	830.0	60.82	888.7	75.71	937.9
7.014	027.0										
0.7760#	520 9**	0 22	614.0	01 00	343 690 0	.15 K 35 /6	763.9	50.97	830.2	65 79	885.9
4.888	579.5	9.33	633 7	25.58	714.0	40.86	787.8	55.70	848.6	70.60	902.7
7,135	597.5	16.32	660.9	30.99	741.9	45.91	809.4	60.97	868.6	75.14	918.0
					959	15 K					
0.4898*	506.4**	8.29	575.9	20.75	661.8	40.88	766.7	55.03	826.4	70.37	883.5
2.961	529.3	11.02	596.9	25.37	688.8	45.63	787.5	60.70	848.4	75.12	899.9
5.165	549.6	14.87	624.4	36.68	747.2	50.42	807.8	65.02	864.4		
					363	.15 K					
0.6254**	473.4**	9.74	559.2	25. 96	668.8	40.72	744.5	55.48	808.2	70.07	864. 0
4.784	515.3	15.62	603.5	31.07	696.3	45.67	767.1	60.25	827.3	75.30	882.1
6.932	535.1	20.86	638.3	35.45	719.3	50.61	788.5	65.38	846.9		
					373	.15 K					
0.7871*	440.9**	11.97	548.7	26.24	647.7	40.70	723.9	55.60	790.0	70.60	847.7
7.473	509.4	15.76	578.1	31.05	674.7	45.40	745.8	60.93	811.3	75 .4 0	864.4
9.20	029.0	20.30	010.0	30.37	097.5	50.36	108.0	65.87	830.1		

^a Values marked with an asterisk are vapor pressures p_s , from ref 12 for CCl₃F and ref 13 for C₂HCl₂F₃. Those values marked with a double asterisk are extrapolated values at the given p_s .



Figure 2. Temperature dependence, T, of ultrasonic speed, u. CCl₃F: ⊖,-- (ref 14), saturated vapor pressure; ⊖, 30 MPa; ⊖, 60 MPa. C2HCl2F3: 0, saturated vapor pressure; 0, 30 MPa; 0, 60 MPa.

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frequently observed, especially in the high-temperature region. This behavior, which appeared as a narrow pulse width in the received signal, gives rise to irreguralities in the experimental results. Lainez et al. (15) also measured the ultrasonic speed in liquid CCI₂F using a pulse echo overlap method at temperatures from 353 to 413 K and pressures up to 200 MPa. When the values at saturation were recalculated by using their 12 experimental points up to about 75 MPa at 353.25 K, the value of 552.8 m·s⁻¹ was obtained, which is low compared with those presented here and reported by Chavez et al. (14). $u_{a_{1}}$ values for C₂HCl₂F₃ differ by about 50 m·s⁻¹ from the value for CCl₃F over the whole temperature range (see Figure 2). With elevated pressure, the difference of u between the two substances, derived from eq 1, tends to decrease.

For these fluids, many sets of accurate values for the saturated liquid densities, $\rho_{p_{1}},$ have been reported. From the extrapolated values of u_p , the isentropic compressibility, κ_s [= $(\rho u^2)^{-1}$] at p_* was estimated by using the ρ_p calculated from equations reported by Downing (12) for CCl₃F and Yokoyama and Takahashi (16) for C2HCl2F3, and the results are shown in Figure 3 as a function of temperature. The saturated liquid densities for both compounds show the same temperature dependence (16), but the values of κ_s for C₂HCl₂F₃ have a larger temperature dependence compared with those for CCI₃F and diverge significantly at higher temperatures. The temperature and pressure dependence of the speed of sound is closely related to the molecular structure in each liquid (17). That is, the speed of sound has a large absolute value for simple molecules having a spherical and/or symmetrical structure, such as CCl₄, as shown in Figure 1. The ultrasonic speed is little influenced by pressure changes, because the molecules are packed to high density (18). For hydoro- and/or chlorofluorocarbons, such as CCI₃F and C₂HCI₂F₃, the van der Waals radius of the florine atom is small compared with that for the

Figure 3. Temperature dependence, *T*, of isentropic compressibility, κ_s , at saturated liquid conditions. CCl₃F: Θ , this work; \Box , estimated from u reported in ref 15. $C_2HCl_2F_3$: 0, this work.

chlorine atom, so these substances will have a larger free volume. The increase of free volume contributes in general to a decrease in the speed of sound and to an increase in the compressibility, and that leads to the strong pressure dependence observed in the case of the nonpolar and weak-polar substances. From these facts, C2HCl2F3 should have a large intermolecular free volume as a result of the complexity of the molecular structure, and the ultrasonic speed and compressibility should be more temperature and pressure dependent than those for CCl₃F.

C₂HCl₂F₃, which is assumed to have to be short atmospheric lifetime, has been recommended as a replacement compound of CCI_aF, since both substances have similar physicochemical properties, such as the critical constants and vapor pressure. However, the present results for the ultrasonic speeds in C₂H-Cl₂F₃ differs considerbly from the values for CCl₃F, especially at higher temperatures. This difference may present some thermodynamic problems in using C₂HCl₂F₃ as a replacement as an alternative compound of CCI₃F.

Registry No. CCl₃F, 75-69-4; C₂HCl₂F₃, 306-83-2.

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PVT and Vapor Pressure Measurements on 1,1-Dichloro-2,2,2-trifluoroethane (HCFC-123)

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A complete set of PVT properties, vapor pressures, and critical parameters of 1,1-dichloro-2,2,2-trifluoroethane (HCFC-123) were measured by a constant-volume method. Values of 134 PVT property data were obtained along 19 isochores in a range of temperatures from 311 to 523 K, pressures from 0.5 to 12 MPa, and densities from 95 to 1440 kg·m⁻³, respectively. Results for 68 vapor pressures were obtained at temperatures from 308 K to the critical temperature, while a vapor pressure correlation representing the experimental data within $\pm 0.26\%$ was developed. The critical pressure was determined as 3.6655 \pm 0.0030 MPa in the course of developing the vapor pressure correlation. The uncertainties of measurements are less than ± 10 mK in temperature, ± 2 kPa in pressure, and ± 0.01 to $\pm 0.14\%$ in density, respectively. The purity of samples used was 99.8 and 99.82 wt %.

Introduction

1,1-Dichloro-2,2,2-trifluoroethane (HCFC-123, $CHCl_2CF_3$) is one of the promising substitutes for the conventional refrigerant CFC-11 (CCl_3F), but studies of the thermodynamic properties of HCFC-123 are rather limited, not only in the quantity of the reported data but also in the range of the measurements up to the present.

In the present study, the vapor pressures and *PVT* properties are reported in extensive temperature, pressure, and density range covering the entire fluid phase. A vapor pressure correlation and the critical pressure have also been derived from the experimental data.

Experimental Apparatus and Procedure

A constant-volume method was applied for measuring the vapor pressures and PVT properties of HCFC-123. Figure 1 shows the experimental apparatus, which is composed of a constant-volume cell immersed in a thermostated bath, a temperature-measurement device, a PID temperature controller, and a pressure-measurement device, etc. We have already measured PVT properties of 1,1,1,2-tetrafluoroethane (HFC-134a) (1), and perfluoro-2-methylpentane (2), with this apparatus.

The inner volume of the constant-volume cell was carefully calibrated by using the density of water and its volume change

Table I.	Uncertainties	of Data	and F	urities	of	Samples	of
HCFC-12	23						

	series	$\Delta T/$ mK	purity/ wt %	Δρ/ (kg·m⁻³)	purity/ wt %
vapor pressures		±10	±2		99.8/99.82
PVT properties	a b c d e f g h i	±10	±2	± 0.1 ± 0.1 ± 0.2 ± 0.3 ± 0.4 ± 0.5 ± 0.5 ± 0.7 ± 0.7	99.82 99.8 99.82 99.82 99.82 99.8 99.8 9
	j k m n o p q r s			± 0.8 ± 0.8 ± 0.9 ± 0.9 ± 0.9 ± 0.9 ± 0.8 ± 0.8 ± 0.8 ± 0.7 ± 0.4	99.82 99.82 99.82 99.82 99.82 99.82 99.82 99.82 99.82 99.82 99.82 99.82

due to changes in temperature and pressure was calculated by using the thermal expansivity and Young's modulus of stainless steel. The volume was about 139 cm³ at room temperature. The density of the sample was determined from the inner volume of the cell and the mass of the sample confined in the cell, which was measured by a precision balance. The uncertainty of the cell volume was about ± 0.02 to $\pm 0.12\%$, depending on temperature and pressure, while the uncertainty of the sample mass was ± 3 mg.

The temperature of silicon oil filled in a thermostated bath was controlled within ± 3 mK by the PID temperature controller. At thermal equilibrium, which was confirmed by the pressure of the sample fluid in the cell being stable, the pressure and the temperature of the sample were measured.

The pressure of the sample was measured by the pressure gauges O1, O2, and O3 in Figure 1. The sample cell is connected with a differential pressure detector (B), which transfers the pressure of the sample to pressure-transfer medium, nitrogen gas. The mechanical behavior of the differential pressure detector (B) was calibrated before and after each series of measurements.

The temperature was measured with a standard platinum resistance thermometer (D in Figure 1), which was located near the sample cell. The platinum resistance thermometer was calibrated according to IPTS-68 at the National Laboratory of Metrology, Ibaraki, Japan.

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