Measurements of the Surface Tension of Four Halogenated Hydrocarbons, CCl_3F, CCl_2F_2, $C_2Cl_3F_3$, and $C_2Cl_2F_4$

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By means of the capillary rise method, we have measured the surface tension of four different kinds of halogenated hydrocarbons, namely, trichlorofluoromethane (CCl_3F; R 11), dichlorodifluoromethane (CCl_2F_2; R 12), trichlorotrifluoroethane ($C_2Cl_3F_3$; R 113), and dichlorotetrafluorethane ($C_2Cl_2F_4$; R 114). Under the coexistence of the sample liquid with its saturated vapor in equilibrium, the measurements have been performed within the maximum uncertainty of 0.12 $mN \cdot m^{-1}$ at temperatures from 273 K up to near the critical point of the respective substances. Under the same experimental conditions, two sets of surface tension data have been obtained with two different Pyrex glass capillaries whose inner radii were 0.1536 ± 0.0004 and 0.1724 ± 0.0005 mm, respectively. The two sets of data were in agreement within 0.1 $mN \cdot m^{-1}$. The data were represented by van der Waals-type correlations with a standard deviation of 0.10 $mN \cdot m^{-1}$ for CCl_3F, 0.04 $mN \cdot m^{-1}$ for CCl_2F_2, 0.08 $mN \cdot m^{-1}$ for $C_2Cl_3F_3$, and 0.07 $mN \cdot m^{-1}$ for $C_2Cl_2F_4$, respectively.

KEY WORDS: Capillary constant; capillary rise method; halogenated hydrocarbons; surface tension.

1. INTRODUCTION

The insufficiency of the experimental surface tension data with respect to technically important fluids has been pointed out by many authors, especially by those who compile the thermodynamic properties of these fluids. In addition, the temperature dependence of the surface tension has not been accurately determined, and the relation between the surface tension and the chemical structure has not been completely resolved.

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The availability of the surface tension data for the halogenated hydro-carbons, which are generally known as various fluorocarbon refrigerants, is not an exceptional case, and the needs for such data have been pointed out in many fields of applications. Earlier (up to 1968) measurements of the surface tension of these substances were limited to four investigations reported by Hovorka and Geiger [1], Lainé [2], Steinle [3], and Grigull and Straub [4]. Since then, several investigators—Dorokhov et al. [5–7], Sinitsyn et al. [8], Heide [9], Muratov and Skripov [10], Watanabe and co-workers [11,12], and Rathjen and Straub [13]—have measured the surface tension of the halogenated hydrocarbons for wider temperature ranges. In spite of all these, additional experimental data are still required.

When the experimental values are not available, the surface tension values may be estimated using Sugden's method. Such an approach has been adopted in the preparation of some of the tables presented in the literature. However, the uncertainty of the reported values in such tables is frequently greater than 5%.

The purpose of the present investigation is to obtain accurate experimental surface tension data for four halogenated hydrocarbons, CCl_3F, CCl_2F_2, $C_2Cl_3F_3$, and $C_2Cl_2F_4$, over wide ranges of temperature and, in addition, to develop correlation schemes in order to represent the experimental data as functions of temperature. Special attention has been given to $C_2Cl_2F_4$, since no experimental results have been reported for this substance in the literature.

2. EXPERIMENTAL

The capillary rise method, well known as one of the most accurate methods, was used in the present study. The schematics of the experimental apparatus are shown in Fig. 1. In order to obtain two sets of surface tension data under the same experimental conditions, two Pyrex glass capillaries (C) were installed vertically in a cell (A) made of either Pyrex glass or glass ceramic. In other words, two independent measurements were made for respective capillaries, without detecting the difference in the vertical locations between two menisci of both capillaries.

The cell was evacuated to 10 Pa or less by a vacuum pump (O). Subsequently, after repeating the flushing and the evacuation of the sample into and from the cell, a prescribed amount of the sample was confined in the cell from a sample bottle (N). The cell was placed in the thermostated bath B of 22 dm^3 in its inner capacity.

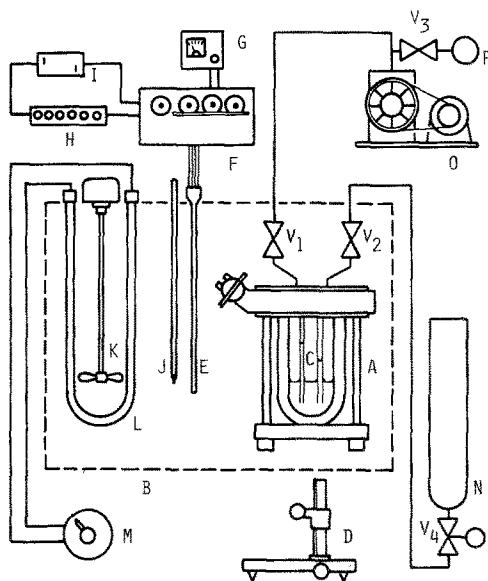As the thermostated fluid, water was used for temperatures of 273 to

Fig. 1. Schematic diagram of the experimental apparatus for the surface tension measurements of the halogenated hydrocarbons. (A) Pyrex glass or glass ceramic cell; (B) thermostated bath; (C) Pyrex glass capillaries; (D) traveling microscope; (E) platinum resistance thermometer; (F) precision double bridge; (G) galvanometer; (H) variable resistor; (I) battery; (J) mercury thermometer; (K) stirrer; (L) electric heater; (M) transformer; (N) sample gas bottle; (O) vacuum pump; (P) vacuum pressure gage; (V_1, V_2, V_3, V_4) valves.

368 K, whereas paraffin oil was used for temperatures of up to 468 K. The temperature of the thermostated fluid was controlled by an electric heater (L) and was maintained constant within ± 10 mK for the sufficiently long duration. After the complete thermal equilibrium was attained, the temperature of the sample in the cell was considered equal to that of the thermostated fluid. The temperature was measured with a platinum resistance thermometer (E) which was installed near the cell in the bath. The thermometer was calibrated precisely at the National Research Laboratory of Metrology, Japan, based on the International Practical Temperature Scale (1968) and the accuracy was confirmed within ± 5 mK.

The location of the meniscus level from the confined liquid surface was measured individually along two different capillaries through the window of

the thermostated bath with a traveling microscope (D) with 10-μm precision in the reading. Thus the observed capillary rise height was determined as the average of three measurements, which agreed well within 30 μm in the worst case.

It should be noted that all of the measurements were carried out under the coexistence of the sample liquid with its saturated vapor in equilibrium.

The two kinds of cells used in the present study are shown in Fig. 2 in detail. Cell I was made of Pyrex glass with Teflon packing, since it is essential to avoid the contamination of the high-purity sample. Two valves connected on the top of the cell were also made of Pyrex glass with Teflon packings for sealing. The inner diameter of cell I was 38 mm and wide enough to obtain the correct liquid level with the sample like halogenated hydrocarbons. Cell I was used in the earlier series of measurements, which were carried out at lower temperatures (below 368 K). When the saturated vapor pressure of the sample was over 2.2 MPa, the Pyrex glass valves were not capable of standing the pressure. Hence cell II was designed for measurements at higher pressures. The bottom half of the cell, as shown in Fig. 2, was made of glass ceramic, whereas the top lid with valves was made of stainless steel SUS 304
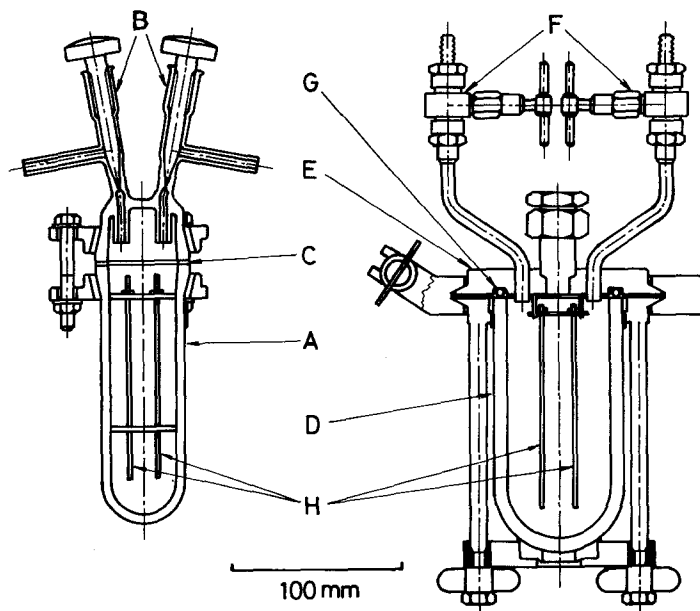

Fig. 2. Cross sections of cell I and cell II. (A) Cell I made of Pyrex glass; (B) Pyrex glass valves; (C) Teflon packing; (D) cell II made of glass ceramic; (E) stainless-steel lid; (F) stainless-steel valves; (G) "Viton" or silicone "O" ring; (H) Pyrex glass capillaries.

with either a "Viton" or a silicone "O" ring. Since glass ceramic has a higher strength and better heat and acid resistance than ordinary glass, cell II could be used for temperatures of up to 468 K and pressures of up to 3.1 MPa, in the critical region of the halogenated hydrocarbons of present interest.

The Pyrex glass capillaries used in the present study were chosen from about 60 capillaries especially manufactured in order to find the most appropriate ones with small and uniform inner bores. The inner radii of the selected two capillaries were determined by means of mercury thread. About 20 mg of mercury was confined in each capillary and the length (about 30 mm) of the mercury thread was measured with a precision of 5 μm in reading. These measurements were conducted under a constant temperature of 293.15 $\pm$ 0.5 K and even a minute difference in the inner radius variation along the capillary length was observed by traveling the mercury thread consecutively. The mass of the mercury thread was measured with a precision of 0.1 mg. A value of 13545.88 kg $\cdot$ m^{-3} [14] was used for the density of mercury at 293.15 K. The final values of the radii were the averages of three series of measurements which agreed within the estimated uncertainty of 0.27%. The length of the two capillaries was about 120 mm, and the greatest variation in the radii along the capillary length was 3.2 μm for the small-bore capillary I and 2.8 μm for the large-bore capillary II. The variations in the radii corresponding to the utilized parts of the capillaries were in the range of 0.1526 to 0.1550 mm for capillary I and in the range of 0.1711 to 0.1725 mm for capillary II. Therefore, if one averaged value of radius was used for one capillary, the uncertainty of the averaged value would be about 1%. However, as mentioned previously, the distribution of the variations of the inner radius was calibrated in advance, and thus the calculations of the surface tension could be done using the exact inner radius corresponding to the location of the sample liquid meniscus.

Prior to the measurements, much attention was paid to the preservation of the high purity of the sample. The halogenated hydrocarbon samples were furnished by Mitsui Fluorochemicals Co., Tokyo, and their purities were 99.99 mol% for both CCl$_3$F and CCl$_2$F$_2$ and 99.9 mol% for both C$_2$Cl$_3$F$_3$ and C$_2$Cl$_2$F$_4$.

All parts of the experimental apparatus that were in contact with the sample were carefully cleaned before the assembly. The parts made of glass were cleaned with hot sulfuric and chromic acids and distilled water, whereas the parts made of stainless steel were cleaned with 20% nitric acid solution and distilled water. Moreover, after assembling the apparatus, the cell was cleaned with the sample liquid itself. Especially in the case of capillaries, their inner walls were carefully cleaned many times with hot sulfuric and chromic acids and distilled water, after which the capillaries were placed in boiling distilled water for 2 or 3 hr.

3. RESULTS

The surface tension, σ, of the four different halogenated hydrocarbons were determined by using the well-known relation given by Rayleigh [15]:

$$\sigma = r(h_0 + r/3 - 0.1288\, r^2/h_0 + 0.1312\, r^3/h_0^2) \times g(\rho' - \rho'')/2 \cos \theta. \qquad (1)$$

In this expression, σ denotes the surface tension (in $N \cdot m^{-1}$), θ is the contact angle of the liquid–vapor meniscus at the inner surface of the capillary, g is the local gravitational acceleration in Yokohama, i.e., $9.79765 \text{ m} \cdot s^{-2}$, ρ' and ρ'' are the saturated liquid and vapor densities (in $kg \cdot m^{-3}$), r is the inner radius of the capillary (in m), and h_0 denotes the capillary rise height observed (in m) as measured from the liquid level of the bottom of the meniscus in the capillary.

The contact angle θ was considered as 0 rad, since the inner walls of the Pyrex glass capillaries were carefully cleaned and the measurements of the capillary rise height were made under the condition of a receding contact angle.

The values of r were corrected for the linear thermal expansion of the Pyrex glass capillary; the greatest contribution of this correction was 0.06% at 468 K.
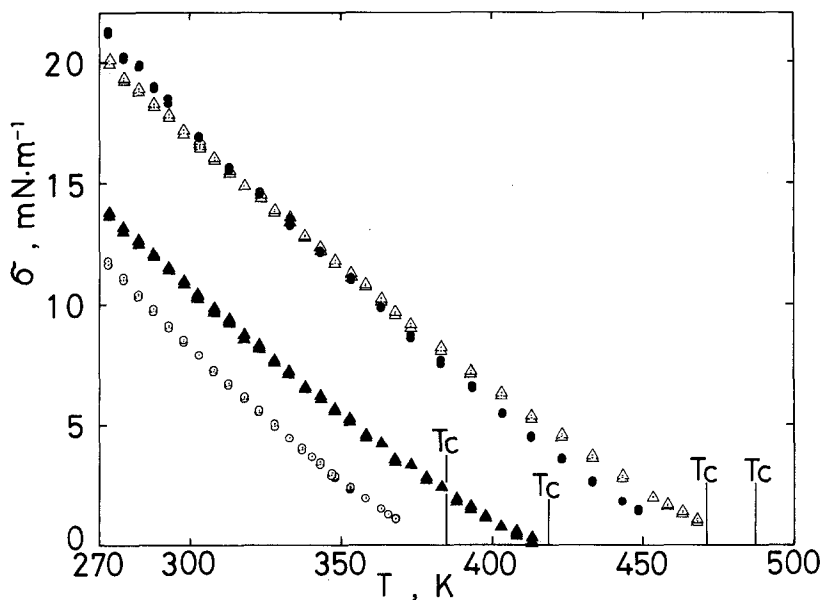

Fig. 3. Temperature dependence of the surface tension σ of four halogenated hydrocarbons. ($\bullet$) CCl_3F; ($\odot$) CCl_2F_2; ($\triangle$) $C_2Cl_3F_3$; ($\blacktriangle$) $C_2Cl_2F_4$.

The saturated liquid and vapor density values of the four halogenated hydrocarbons have been taken from the thermodynamic correlations [16]. However, in the case of $C_2Cl_3F_3$, the density values for temperatures over 373 K were obtained from the temperature-density diagram based on the experimental values of Benning and McHarness [17], since the correlations in the Ref. 16 were applicable only for temperatures of up to 373 K.

Prior to the measurements on the halogenated hydrocarbons, the surface tension of distilled water was measured at 298.15 K using the same experimental apparatus. The result was 71.89 mN $\cdot$ m^{-1}, which is in good agreement with the value of 71.99 $\pm$ 0.36 mN $\cdot$ m^{-1} for light water recommended recently by the IAPS [18].

The results obtained for the four halogenated hydrocarbons are given in Table I and Fig. 3. The temperature ranges covered are 273 to 448 K for CCl_3F, 273 to 368 K for CCl_2F_2, 273 to 468 K for $C_2Cl_3F_3$, and 273 to 413 K for $C_2Cl_2F_4$, respectively.

Based on the results for the surface tension, the capillary constant values were calculated from the following relation:

$$a^2 = r(h_0 + r/3 - 0.1288\, r^2/h_0 + 0.1312\, r^3/h_0^2) \times g/g_n,$$

$$= 2\sigma/[g_n(\rho' - \rho'')], \tag{2}$$

where g_n denotes the normal gravitational acceleration of 9.80665 m $\cdot$ s^{-2}. The values of a^2 are also given in Table I.

As a part of the error analysis of the experimental data, various factors affecting the parameters in Eq. (1) were studied. The estimated uncertainties with respect to the inner radii of capillaries were 0.27% whereas those of the liquid and vapor density values were considered to be less than 0.2%. With increasing temperature, the capillary rise height decreased from approximately 19 to 2 mm, and hence the uncertainties in the height reading were estimated to be in the range of 0.1 to 1%. Therefore the uncertainties in the experimental surface tension values were estimated to be 0.6% for low temperatures and 1 to 2% for high temperatures. Expressed in absolute values, the uncertainties were 0.1 mN $\cdot$ m^{-1} for low temperatures and 0.01 mN $\cdot$ m^{-1} for high temperatures.

For the purpose of correlating the temperature dependence of the surface tension of the four halogenated hydrocarbons shown in Fig. 3, the following van der Waals-type expression may be used:

$$\sigma = \sigma_0(1 - T/T_c)^n, \tag{3}$$

where T and T_c denote the temperature (in K) and the critical temperature

Table I. Experimental Values of the Surface Tension σ and the Capillary Constant a^2 of CCl_3F, CCl_2F_2, $C_2Cl_3F_3$, and $C_2Cl_2F_4$ at Temperature T

	Capillary I		Capillary II			Capillary I		Capillary II	
T (K)	σ (mN·m^{-1})	a^2 (mm^2)	σ (mN·m^{-1})	a^2 (mm^2)	T (K)	σ (mN·m^{-1})	a^2 (mm^2)	σ (mN·m^{-1})	a^2 (mm^2)
				CCl_3F Series I					
273.19	21.28	2.833	21.21	2.824	323.18	14.55	2.118	14.62	2.127
283.18	19.84	2.683	19.78	2.676	333.16	13.29	1.977	13.52	2.010
293.17	18.54	2.551	18.35	2.525	343.18	12.16	1.851	12.19	1.855
303.15	17.12	2.397	17.15	2.401	353.19	11.02	1.720	11.11	1.734
313.20	15.76	2.249	15.88	2.265	363.20	9.84	1.579	9.87	1.583
				CCl_3F Series II					
278.15	20.16	2.705	20.27	2.719	288.14	18.90	2.577	18.96	2.585
				CCl_3F Series III					
333.14	13.21	1.964	13.31	1.979	413.17	4.48	0.878	4.53	0.888
363.15	9.78	1.569	9.89	1.587	423.17	3.54	0.740	3.58	0.747
373.16	8.64	1.430	8.75	1.447	433.17	2.60	0.586	2.66	0.599
383.15	7.56	1.293	7.70	1.318	443.19	1.81	0.453	1.82	0.455
393.17	6.55	1.166	6.61	1.176	448.18	1.40	0.376	1.44	0.387
403.17	5.49	1.021	5.52	1.027					
				CCl_2F_2 Series I					
273.18	11.66	1.725	11.81	1.747	318.16	6.12	1.067	6.18	1.077
278.14	11.03	1.655	11.10	1.666	323.14	5.53	0.989	5.62	1.005
283.14	10.37	1.580	10.42	1.587	328.14	4.94	0.909	5.08	0.934
288.14	9.76	1.509	9.81	1.518	333.15	4.39	0.833	4.49	0.851
293.15	9.10	1.431	9.16	1.440	337.16	3.98	0.775	4.05	0.788
298.15	8.51	1.363	8.55	1.370	340.14	3.72	0.741	3.73	0.742
303.17	7.90	1.289	7.94	1.296	343.14	3.37	0.687	3.41	0.695
308.15	7.30	1.215	7.36	1.225	347.15	2.95	0.622	2.97	0.627
313.15	6.68	1.136	6.78	1.155					
				CCl_2F_2 Series II					
303.15	7.93	1.294	7.87	1.285	348.16	2.88	0.613	2.84	0.605
313.14	6.67	1.136	6.70	1.141	353.16	2.37	0.530	2.34	0.524
323.17	5.55	0.992	5.52	0.987	358.16	1.93	0.459	1.92	0.455
333.15	4.43	0.839	4.43	0.841	363.16	1.49	0.381	1.46	0.373
343.15	3.41	0.695	3.38	0.689	366.16	1.26	0.339	1.26	0.338
343.16	3.43	0.699	3.37	0.688	368.17	—	—	1.08	0.302
				$C_2Cl_3F_3$ Series I					
298.19	17.24	2.252	17.04	2.226	338.14	12.80	1.796	12.85	1.803
308.14	16.04	2.129	15.99	2.123	348.16	11.66	1.670	11.78	1.688
318.15	14.95	2.019	14.89	2.011	358.19	10.75	1.575	10.79	1.581
328.14	13.95	1.919	13.87	1.908	368.15	9.58	1.438	9.68	1.454

Table I. Continued

C₂Cl₃F₃ Series II

278.14	19.29	2.446	19.33	2.451	288.15	18.12	2.342	18.15	2.334

C₂Cl₃F₃ Series III

273.17	19.92	2.507	20.10	2.530	303.15	16.49	2.171	16.58	2.183
283.15	18.81	2.402	18.91	2.415	313.13	15.41	2.063	15.51	2.076
293.14	17.75	2.301	17.85	2.314	323.14	14.52	1.978	14.50	1.977

C₂Cl₃F₃ Series IV

333.14	13.36	1.856	13.51	1.876	353.15	11.23	1.626	11.26	1.632
343.11	12.22	1.731	12.29	1.743	363.17	10.23	1.517	10.25	1.520

C₂Cl₃F₃ Series V

303.16	16.60	2.185	16.74	2.203	413.17	5.29	0.918	5.38	0.933
323.15	14.35	1.956	14.52	1.979	423.18	4.49	0.817	4.58	0.832
343.14	12.16	1.723	12.32	1.746	433.18	3.60	0.693	3.66	0.704
363.14	10.08	1.494	10.25	1.520	443.18	2.81	0.578	2.87	0.591
373.15	9.13	1.390	9.17	1.395	453.18	2.00	0.448	2.01	0.452
383.15	8.08	1.265	8.25	1.291	458.18	1.64	0.389	1.65	0.390
393.16	7.16	1.155	7.24	1.167	463.19	1.37	0.346	1.32	0.331
403.16	6.19	1.032	6.34	1.058	468.10	1.01	0.276	1.07	0.292

C₂Cl₂F₄ Series I

273.20	13.67	1.832	13.80	1.848	313.14	9.34	1.375	9.34	1.376
283.15	12.54	1.715	12.65	1.730	323.13	8.23	1.250	8.27	1.256
293.15	11.45	1.601	11.52	1.611	333.14	7.17	1.127	7.19	1.129
303.14	10.29	1.475	10.38	1.489	343.15	6.14	1.002	6.15	1.003

C₂Cl₂F₄ Series II

278.15	13.03	1.763	13.18	1.783	318.15	8.59	1.283	8.77	1.311
288.15	11.99	1.658	12.07	1.669	328.14	7.61	1.174	7.64	1.179
298.14	10.88	1.540	10.94	1.549	338.16	6.53	1.044	6.61	1.058
308.14	9.71	1.411	9.78	1.421					

C₂Cl₂F₄ Series III

303.14	10.34	1.483	10.45	1.499	373.15	3.35	0.642	3.33	0.638
313.15	9.25	1.363	9.39	1.383	383.16	2.41	0.501	2.43	0.504
323.14	8.17	1.240	8.29	1.259	393.16	1.65	0.382	1.53	0.354
333.15	7.14	1.122	7.25	1.139	403.16	—	—	0.77	0.209
343.14	6.12	0.999	6.21	1.013	408.17	—	—	0.54	0.168
353.15	5.13	0.875	5.27	0.899	413.17	—	—	0.16	0.063
363.15	4.22	0.759	4.23	0.760					

C₂Cl₂F₄ Series IV

308.15	9.75	1.417	9.82	1.427	388.16	1.94	0.424	1.92	0.419
328.14	7.62	1.176	7.69	1.187	398.15	1.17	0.291	1.13	0.281
348.16	5.57	0.928	5.70	0.950	403.21	0.80	0.217	0.75	0.204
358.15	4.61	0.806	4.65	0.814	408.16	0.50	0.154	0.42	0.131
368.15	3.62	0.671	3.66	0.678	413.17	0.27	0.103	—	—
378.15	2.74	0.545	2.79	0.555					

Table II. Numerical Constants n and σ_0 and the Critical Temperature T_c in the Present Correlations; $\sigma = \sigma_0(1 - T/T_c)^n$

Substance	σ_0 (mN · m^{-1})	n	T_c (K)
CCl$_3$F	62.247	1.2525	471.15[16]
CCl$_2$F$_2$	55.854	1.2624	384.95[25]
C$_2$Cl$_3$F$_3$	55.378	1.2382	487.25[16]
C$_2$Cl$_2$F$_4$	50.844	1.2363	418.85[16]

(in K), respectively, and σ_0 and n are numerical constants which depend only on the substance. The data were fitted by Eq. (3) using the least-squares method, which yielded the following standard deviations: 0.10 mN · m^{-1} for CCl$_3$F, 0.04 mN · m^{-1} for CCl$_2$F$_2$, 0.08 mN · m^{-1} for C$_2$Cl$_3$F$_3$, and 0.07 mN · m^{-1} for C$_2$Cl$_2$F$_4$. The numerical constants thus determined for the four halogenated hydrocarbons are given in Table II. The values of the critical temperature and their literature sources are also given in Table II.

The deviations of the present results for the present correlations are
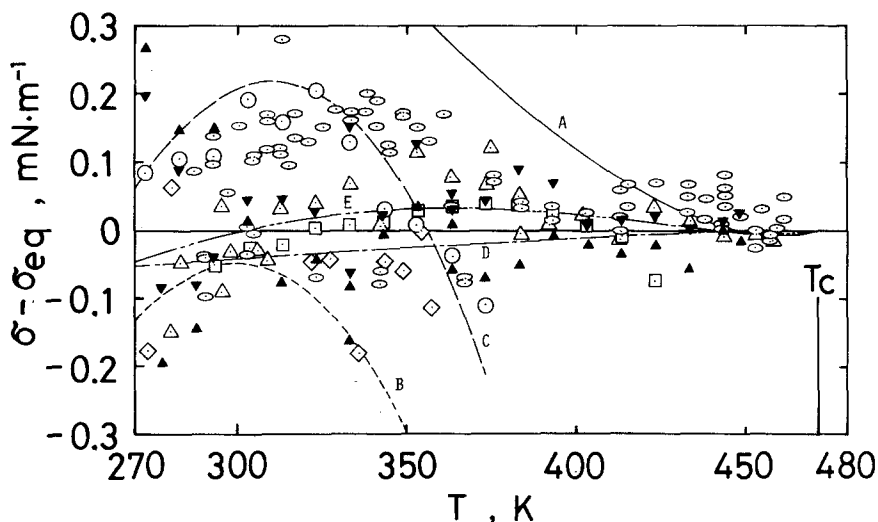
Fig. 4. Deviations of the present and previously reported surface tension values σ of CCl$_3$F from the present correlation's values σ_{eq} for the temperatures from 273 K up to the critical point. (▲) Present work, capillary I; (▼) present work, capillary II; (◆) Dorokhov et al. [5–7]; (▣) Sinitsyn et al. [8]; (⊙) Heide [9]; (◌) Watanabe and Watanabe [11]; (△) Rathjen and Straub [13]; (curve A) general correlation of Dorokhov et al. [5–7]; (curve B) linear correlation of Dorokhov et al. [5–7]; (curve C) linear correlation of Heide [9]; (curve D) van der Waals-type correlation of Rathjen and Straub [13]; (curve E) modified van der Waals-type correlation of Rathjen and Straub [13].
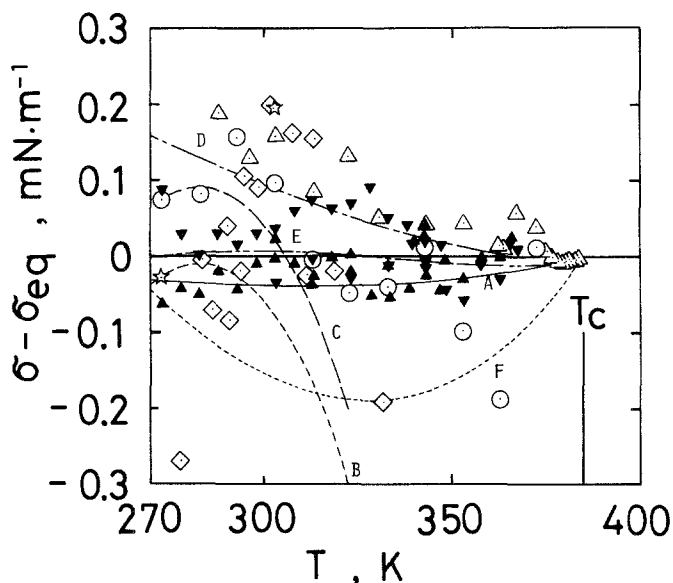
Fig. 5. Deviations of the present and previously reported surface tension values σ of CCl_2F_2 from the present correlation's values σ_{eq} for the temperatures from 273 K up to the critical point. (▲) Present work, capillary I; (▼) present work, capillary II; (☆) Lainé [2]; (◊) Dorokhov et al. [5–7]; (⊙) Heide [9]; (△) Rathjen and Straub [13]; (curve A) general correlation of Dorokhov et al. [5–7]; (curve C) linear correlation of Heide [9]; (curve D) van der Waals-type correlation of Rathjen and Straub [13]; (curve E) modified van der Waals-type correlation of Rathjen and Straub [13]; (curve F) van der Waals-type correlation of Watson [19].

shown in Fig. 4 for CCl_3F, in Fig. 5 for CCl_2F_2, in Fig. 6 for $C_2Cl_3F_3$, and in Fig. 7 for $C_2Cl_2F_4$. Also in Figs. 4–7 deviations of the previously reported experimental data [1,2,5–9,11,13] and proposed correlations [5–7,9,13,19] from the present correlations are included.

4. DISCUSSION AND CONCLUSION

The reproducibility of the present measurements of the surface tension has been confirmed by the following two points. First, most of the two sets of data observed with different capillaries are in good agreement with each other, within ± 0.1 mN $\cdot$ m^{-1}. Second, most of the data obtained during different series of experiments are also in good agreement, within ± 0.1 mN $\cdot$ m^{-1}.

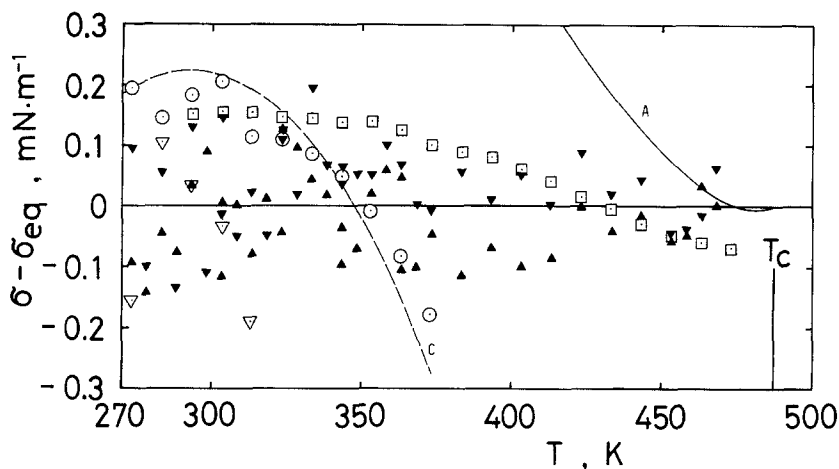The established van der Waals-type correlations are considered to be

Fig. 6. Deviations of the present and previously reported surface tension values σ of $C_2Cl_3F_3$ from the present correlation's value σ_{eq} for the temperatures from 273 K up to the critical point. (▲) Present work, capillary I; (▼) present work, capillary II; (∇) Hovorka and Geiger [1]; (□) Sinitsyn et al. [8]; (⊙) Heide [9]; (curve A) general correlation of Dorokhov et al. [5–7], (curve C) linear correlation of Heide [9].

satisfactory, because the standard deviations of the present data from these correlations are less than the estimated experimental uncertainties. The values of the exponent n in the present correlations are found to be in the range of 1.24 to 1.26 for each substance. These values are somewhat greater than the 1.22 proposed by Guggenheim [20] and somewhat smaller than the 1.28 given by Rathjen and Straub [13].

By using deviation plots such as those given in Figs. 4–7, the previously reported data can be compared with the present data for the temperature range from 273 K up to the critical temperature. The experimental data by Sinitsyn et al. [8] for CCl_3F and $C_2Cl_3F_3$, by Watanabe and Watanabe [11] for CCl_3F, and by Rathjen and Straub [13] for CCl_3F and CCl_2F_2 are in good agreement with the present results, within ± 0.2 mN · m^{-1} over the entire temperature ranges. The results reported by Heide [9] for CCl_3F, CCl_2F_2, and $C_2Cl_3F_3$ at temperatures below 373 K are in reasonably good agreement with the present results, within ± 0.2 mN · m^{-1}, but the temperature dependence of his data is somewhat different. The data of Dorokhov et al. [5–7] for CCl_3F and CCl_2F_2 have a large scatter and hence some of them are not given in the figures. The data of Lainé [2] for CCl_2F_2 and of Hovorka and Geiger [1] for $C_2Cl_3F_3$ at low temperatures are in good agreement with the present data, within ± 0.2 mN · m^{-1}. The results given by Steinle [3] for CCl_2F_2 are very low, thus they are not included in Fig. 5.
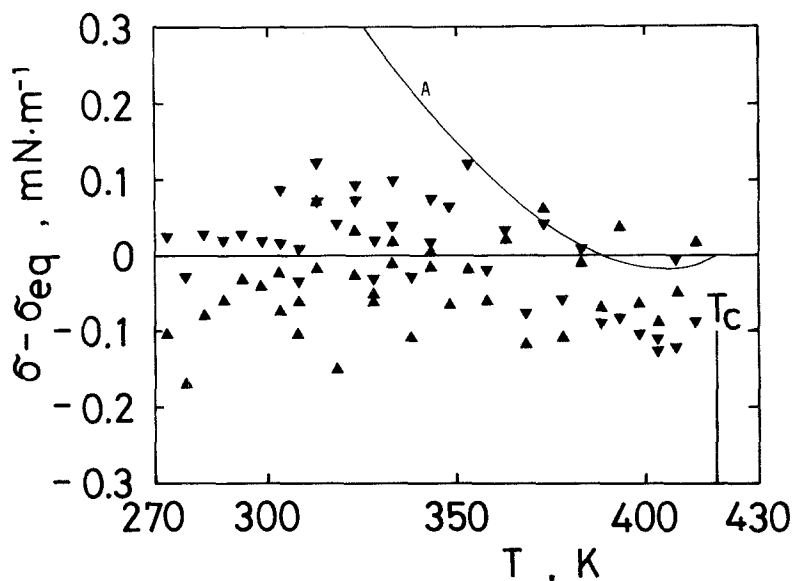
Fig. 7. Deviations of the present and previously reported surface tension values σ of $C_2Cl_2F_4$ from the present correlation's values σ_{eq} for the temperatures from 273 K up to the critical point. (▲) Present work, capillary I; (▼) present work, capillary II; (curve A) general correlation of Dorokhov et al. [5–7].

The compiled values of Vargaftik [21] for CCl_2F_2, of Danilova et al. [22,23] for CCl_3F, CCl_2F_2, and $C_2Cl_3F_3$, and by Tomanovskaya and Koltova [24] for CCl_3F, CCl_2F_2, $C_2Cl_3F_3$, and $C_2Cl_2F_4$ were not included in the figures because of their large deviations.

Other than the two large values (at 298.15 K) compiled by Tomanovskaya and Koltova, no results for $C_2Cl_2F_4$ had been reported previously in the literature.

One may conclude that the data presented in this paper are in good agreement with most of the previously reported results, generally within ±0.2 mN $\cdot$ m^{-1}.

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