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Experimental Surface Tensions for HFC-32, HCFC-124, HFC-125, HCFC-141b, HCFC-142b, and HFC-152a[1]

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The surface tension of six alternative refrigerants, i.e., HFC-32 (CH_2F_2), HCFC-124 ($CHClFCF_3$), HFC-125 (CHF_2CF_3), HCFC-141b (CH_3CCl_2F), HCFC-142b (CH_3CClF_2), and HFC-152a (CH_3CHF_2), has been measured in the present study. The measurements were conducted under equilibrium conditions between the liquid and its saturated vapor. The differential capillary-rise method (DCRM) used two glass capillaries, with inner radii of 0.3034 ± 0.0002 and 0.5717 ± 0.0002 mm, respectively. Temperatures in the range from 270 to 340 K were considered. The accuracy of surface tension measurements is estimated to be within ± 0.2 $mN \cdot m^{-1}$. The temperatures are accurate to within ± 20 mK. The temperature dependence of the resultant data were successfully represented by van der Waals' correlations to within ± 0.1 $mN \cdot m^{-1}$ for each substance. Available surface tension data are compared with the present data.

KEY WORDS: differential capillary-rise method; HFC-32; HCFC-124; HFC-125; HCFC-141b; HCFC-142b; HFC-152a; refrigerants; surface tension.

1. INTRODUCTION

The deadline for phasing out the production of CFCs was moved up by the Parties to the Montreal Protocol at their Copenhagen meeting in November 1992. It was also decided to make HCFCs controlled substances with a virtual phase-out by the year 2020. Therefore, updated and accurate knowledge of the thermophysical properties of alternative refrigerants are

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required. Surface tension is a basic thermophysical property with respect to a vapor–liquid interface and is required to analyze the heat transfer during boiling and condensation in refrigeration and heat pump systems. This work presents the surface tension measurements for six CFC alternatives, that is, three HCFCs (HCFC-124, HCFC-141b, and HCFC-142b) and three HFCs (HFC-32, HFC-125, and HFC-152a). There is intense current interest in the three particular HFCs included above and their mixtures as replacements for HCFC-22 in air-conditioners and heat pumps.

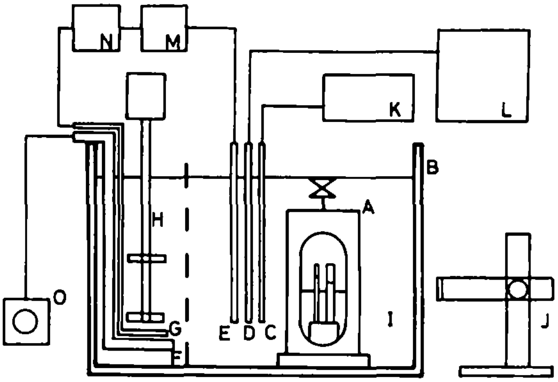
2. EXPERIMENTS

The experimental technique employed in obtaining surface tension data is based on the capillary-rise method, which is well-known to be one of the most accurate methods. The single capillary-rise method (SCRM) was used in our earlier works [1, 2] for some CFCs and HCFCs. However, the differential capillary-rise method (DCRM) is adopted in our recent work [3] for both HCFC-123 and HFC-134a, so as to avoid the difficult measurement of the bulk meniscus level (except for making a small correction). In the present measurements, the differential capillary-rise method is adopted. In this method, two different capillaries with larger and smaller radii r_1 and r_2 are used. The difference in the height (h_1–h_2) between the menisci in two capillaries is derived using Rayleigh's correction [4] for the value. Then, the surface tension σ is determined by the following relation:

$$\sigma = \frac{g(\rho' - \rho'')(h_1 - h_2)}{2(1/r_1 - 1/r_2)\cos\theta} \qquad (1)$$

where θ denotes the contact angle of the vapor–liquid meniscus at the inner surface of capillary, g is the local gravitational acceleration at Iwaki, Japan ($=9.8002\ \mathrm{m \cdot s^{-2}}$), and ρ' and ρ'' are the saturated liquid and vapor densities, respectively.

A schematic diagram of the experimental apparatus is shown in Fig. 1. Two Pyrex glass capillaries are placed vertically in a pressure vessel composed of a thick-walled Pyrex glass tube (17 mm in inner diameter and 25 mm in outer diameter). The inner radii of two capillaries are 0.3034 ± 0.0002 and 0.5717 ± 0.0002 mm, respectively. These radii were determined by means of a mercury thread. The pressure vessel was installed in the thermostated bath ($400 \times 500 \times 300$ mm in inner dimensions). The temperature in the bath was maintained constant to within ± 5 mK and measured based on the IPTS-68. The measured temperature values were converted to ITS-90.



A : Pressure Vessel
B : Thermostated Bath
C, D : Platinum Resistance Thermometer
E : Thermocouple
F : Primary Heater(1.5 kW)
G : Secondary Heater(300 W)
H : Stirrer
I : Heat Transfer Medium
J : Traveling Microscope
K : Thermometer Bridge
L : Recorder
M : PID Controller
N : Voltage Converter
O : Voltage Transformer

Fig. 1. Schematic diagram of the experimental apparatus for the surface tension measurements of the alternative refrigerants.

The difference in rise heights in the capillaries was determined by the measurement of the location of meniscus in each capillary. The location of the bottom of meniscus was measured through the window of the thermostated bath by a traveling microscope with an uncertainty of ±0.02 mm. The contact angle θ is assumed to be zero, since the inner walls of the capillaries were carefully washed and the capillary-rise height measurements were performed under the condition of a receding contact angle. The saturated liquid and vapor density values are calculated from the correlations based on the available thermodynamic property data for HFC-32 [5, 6], HCFC-124 [7], HFC-125 [5, 8], HCFC-141b [9, 10], HCFC-142b [11–13], and HFC-152a [14, 15].

The purities of the samples are not less than 99.98 wt% for HFC-32, 99.90 wt% for HCFC-124 and 99.93 wt% for HCFC-141b, and 99.99 wt% for HFC-125, HCFC-142b, and HFC-152a.

All of the measurements were carried out under equilibrium conditions between the liquid and its saturated vapor. Through the total error analysis, it is concluded that the accuracies of the measurements for surface tensions and temperatures are estimated to be within $\pm 0.2\ \mathrm{mN \cdot m^{-1}}$ and ±20 mK, respectively.

3. RESULTS AND DISCUSSION

Each experiment was repeated to verify the reproducibility of the measurements. The experimental results are given in Table I. The capillary constant a^2 is also listed in Table I, because it does not include the uncertainty of density values. The relation between the surface tension and the capillary constant is

$$a^2 = \frac{(h_1 - h_2)}{[(1/r_1) - (1/r_2)] \cos \theta} \frac{g}{g_n} = \frac{2\sigma}{g_n(\rho' - \rho'')} \tag{2}$$

where g_n denotes the normal gravitational acceleration of 9.80665 $\mathrm{m \cdot s^{-2}}$.

These results show that the surface tension decreases with increasing temperature and becomes zero at the critical point. The following expression by van der Waals is known to represent the temperature dependence of the surface tension σ well:

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

where T and T_c denote the temperature (K) and the critical temperature (K), respectively. The σ_0 and n are numerical constants which depend on the substance. In our previous paper [16], we confirmed that correlations using Eq. (3) represent the surface tension of 17 different fluorocarbon refrigerants well. Therefore σ_0 and n for alternative refrigerants have been determined based on the present results by least-squares fitting. The values of constants σ_0 and n resulting from the fitting, and of the cited critical temperature T_c together with the literature sources [14, 17–20] for these substances, are listed in Table II. The standard deviations of the present results from the fitted equations are also listed in Table II. For the purpose of comparing surface tension data and correlations and discussing their reliability, the deviation plots from the fitted equations for six refrigerants are illustrated. However, on account of limited space, only the deviation plots for three HFCs are shown in Figs. 2–4.

From the deviation plots, it is clear that the present results are scattered within $\pm 0.1\ \mathrm{mN \cdot m^{-1}}$, which is smaller than the estimated accuracy of $\pm 0.2\ \mathrm{mN \cdot m^{-1}}$, which confirms the reproducibility of measurements. As for the correlation, van der Waals' expression represents well the present results for each substance. The values of the constant n fall between 1.2 and 1.3, which is the usual range for most fluids.

The surface tension data are compared in the deviation plots with those reported by other investigators. No experimental data are available for HCFC-124. For HFC-32 and HFC-125, measurements and correlations by Schmidt and Moldover [21] have been reported previously. Figures 2 and 3 show agreement between their data and the present results to within $0.2\ \mathrm{mN \cdot m^{-1}}$. Our data do not extend to the high-temperature range, but

Table I. Experimental Results on the Surface Tension σ and the Capillary Constant a^2 at Temperature T_{90}

T_{90} (K)	a^2 (mm^2)	σ (mN·m^{-1})
	HFC-32	
273.21	2.170	10.97
278.41	2.035	10.07
283.15	1.906	9.23
288.76	1.764	8.32
293.06	1.648	7.60
298.98	1.487	6.62
303.38	1.384	5.99
308.65	1.243	5.18
313.67	1.114	4.45
318.53	0.966	3.69
323.51	0.823	2.97
328.64	0.699	2.35
333.14	0.541	1.69
	HCFC-124	
275.84	1.796	12.36
279.28	1.751	11.98
283.25	1.687	11.46
288.33	1.622	10.90
293.27	1.545	10.27
298.27	1.461	9.59
303.18	1.397	9.04
308.37	1.320	8.40
313.17	1.249	7.82
318.54	1.197	7.35
323.29	1.107	6.66
328.63	1.056	6.20
333.26	0.966	5.54
338.94	0.920	5.12
343.97	0.836	4.51
	HFC-125	
273.23	1.108	6.94
278.84	1.017	6.20
280.30	0.985	5.96
283.40	0.946	5.63
283.84	0.933	5.54
284.62	0.920	5.44
288.90	0.856	4.93
291.79	0.810	4.58

Table I. *(Continued)*

T_{90} (K)	a^2 (mm^2)	σ ($mN \cdot m^{-1}$)
293.15	0.798	4.47
294.04	0.765	4.26
295.49	0.765	4.21
296.10	0.745	4.08
298.34	0.707	3.81
301.21	0.668	3.51
303.17	0.617	3.19
303.33	0.622	3.22
303.49	0.635	3.28
308.20	0.550	2.71
HCFC-141b		
275.66	3.383	21.06
278.70	3.325	20.61
283.18	3.260	20.06
288.72	3.164	19.30
293.70	3.099	18.75
298.40	3.009	18.06
304.69	2.918	17.33
308.44	2.847	16.79
313.36	2.783	16.27
318.10	2.699	15.64
323.27	2.634	15.11
328.31	2.531	14.37
333.30	2.480	13.93
338.04	2.376	13.21
343.31	2.331	12.81
HCFC-142b		
274.05	2.538	14.42
278.56	2.460	13.84
283.79	2.376	13.21
285.45	2.351	13.02
287.62	2.305	12.70
293.76	2.215	12.01
298.08	2.131	11.43
303.09	2.035	10.76
304.27	2.028	10.69
308.01	1.951	10.17
313.06	1.880	9.65
317.91	1.796	9.07
323.24	1.712	8.49
327.99	1.616	7.88
334.23	1.474	7.01
339.10	1.389	6.47

Table I. *(Continued)*

T_{90} (K)	a^2 (mm^2)	σ (mN·m^{-1})
	HFC-152a	
273.48	2.828	13.17
278.42	2.712	12.46
283.58	2.589	11.72
288.38	2.473	11.03
291.71	2.409	10.63
293.21	2.370	10.41
296.89	2.293	9.94
297.31	2.267	9.82
298.30	2.248	9.70
303.18	2.138	9.06
309.33	1.990	8.23
313.28	1.899	7.73
318.16	1.803	7.18
319.21	1.770	7.02
323.29	1.661	6.45
328.18	1.538	5.82
333.08	1.429	5.26
338.00	1.307	4.66
343.17	1.191	4.09

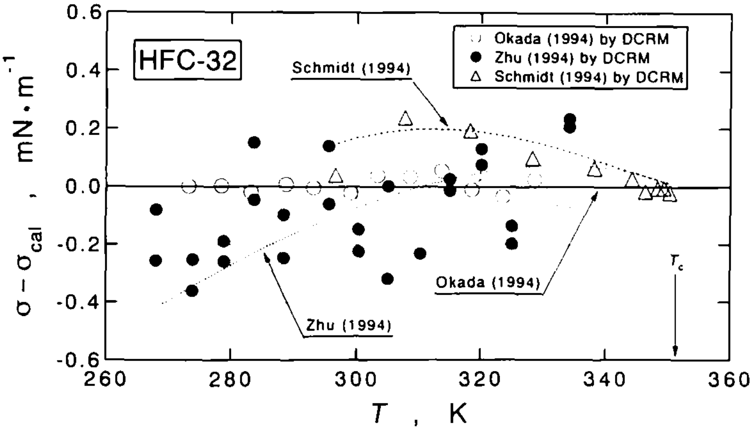


Fig. 2. Deviations of the present and previously reported surface tension values σ of HFC-32 from the calculated values σ_{cal} by the fitted equation.

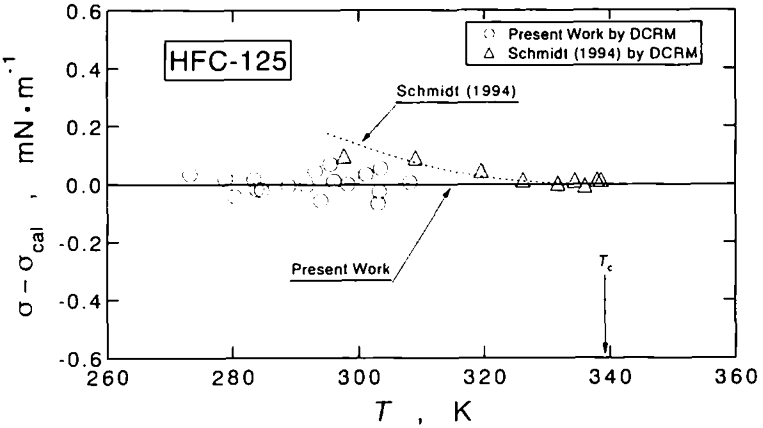


Fig. 3. Deviations of the present and previously reported surface tension values σ of HFC-125 from the calculated values σ_{cal} by the fitted equation.

the present correlations represent the results by Schmidt et al. well at high temperatures. This shows that van-der-Waals type correlations interpolate well between the low-temperature range and the critical temperature. In addition, Zhu and Lu [25] recently reported measurements and correlation of surface tension for HFC-32. Although the scatter of their data is appreciable, those values also agree with the present results as shown in Fig. 2.

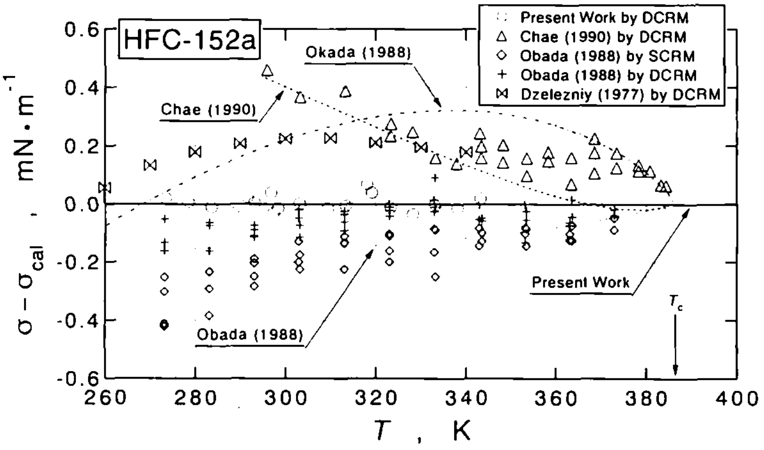


Fig. 4. Deviations of the present and previously reported surface tension values σ of HFC-152a from the calculated values σ_{cal} by the fitted equation.

Table II. Numerical Constants in Eq. (3)

Refrigerant	σ_0 (mN · m^{-1})	n	T_c (K)	SD (mN · m^{-1})
HFC-32	72.16	1.252	351.26[17]	0.03
HCFC-124	52.19	1.203	395.35[18]	0.06
HFC-125	52.60	1.240	339.17[17]	0.04
HCFC-141b	60.87	1.235	477.31[19]	0.06
HCFC-142b	55.14	1.214	410.26[20]	0.06
HFC-152a	59.06	1.221	386.41[14]	0.03

Considerable data are available for HFC-152a, and a comparison with the present correlation is shown in Fig. 4. The results by Chae et al. [22] are larger than the present results and those by Dzelezniy and Krasnoyurchenko [23] are between Chae's data and the present results. The data set reported by Obata [24] are smaller than the present results. However, the present correlation represents most of these data to within ± 0.4 mN · m^{-1}.

For HCFC-141b and HCFC-142b, only one experimental data set by Chae et al. [22] is available. In case of HCFC-141b, the results by Chae et al. are slightly larger than the present results. On the other hand, for HCFC-142b the results by Chae et al. agree with the present results to within 0.2 mN · m^{-1}.

The deviation plots also show that our fittings are successful since the present correlations represent well most experimental surface-tension data for wider temperature ranges.

4. CONCLUSION

The surface tension of six alternative refrigerants was measured over a wide temperature range using the differential capillary-rise method. Based upon the results of this study, a correlation based on van der Waals' expression was developed for each refrigerant.

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REFERENCES

1. K. Watanabe and M. Okada, *Int. J. Thermophys.* **2**:163 (1981).
2. M. Okada, T. Arima, M. Hattori, and K. Watanabe, *J. Chem. Eng. Data* **33**:399 (1988).
3. Y. Higashi, T. Ikeda, T. Kuwana, and M. Okada, *Trans. JAR* **9**:191 (1992) (in Japanese).
4. O. M. Rayleigh, *Proc. Roy. Soc. London Ser. A* **92**:184 (1915).
5. J. V. Widiatmo, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **39**:304 (1994).
6. Z. Y. Qian, A. Nishimura, H. Sato, and K. Watanabe, *JSME Int. J.* **36**:665 (1993).
7. I. R. Shankland, R. S. Basu, and D. P. Wilson, *ASHRAE Trans.* **96**:317 (1990).
8. L. C. Wilson, W. V. Wilding, G. M. Wilson, R. L. Rowley, V. M. Felix, and T. Chisolm-Carter, *Fluid Phase Equil.* **80**:167 (1992).
9. Y. Maezawa, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **36**:151 (1991).
10. D. Arnaud, S. Macaudiere, L. Niveau, and S. Wosinski, *Proc. XVIIIth Int. Cong. Refrig.*, Montréal, Canada (1991), p. 621.
11. Y. Maezawa, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **36**:148 (1991).
12. M. Fukushima and N. Watanabe, *Trans. JAR* **9**:247 (1992) (in Japanese).
13. N. Yada, K. Kumagai, T. Tamatsu, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **36**:12 (1991).
14. Y. Higashi, M. Ashizawa, Y. Kabata, T. Majima, M. Uematsu, and K. Watanabe, *JSME Int. J.* **30**:1106 (1987).
15. T. Tamatsu, T. Sato, H. Sato, and K. Watanabe, *Proc. 13th Japan Symp. Thermophys. Prop.* (1992), p. 45.
16. M. Okada and K. Watanabe, *Heat Transfer Japanese Res.* **17**:35 (1988).
17. Y. Higashi, *Int. J. Refrig.* **17**:524 (1994).
18. M. Fukushima and N. Watanabe, *Trans. JAR* **10**:75 (1993) (in Japanese).
19. L. A. Weber, *Proc. XVIIIth Int. Cong. Refrig.*, Montréal, Canada (1991), p. 616.
20. S. Tanikawa, J. Tatoh, Y. Maezawa, H. Sato, and K. Watanabe, *J. Chem. Eng. Data* **37**:74 (1992).
21. J. W. Schmidt and M. R. Moldover, *J. Chem. Eng. Data* **39**:39 (1994).
22. H. B. Chae, J. W. Schmidt, and M. R. Moldover, *J. Phys. Chem.* **94**:8840 (1990).
23. V. P. Dzelezniy and A. G. Krasnoyurchenko, *Teplofiz. Svoistva Veshchestv i Mater.* **11**:55 (1977) (in Russian).
24. M. Obata, Master thesis (Nagaoka University of Technology, Nagaoka, Japan, 1988) (in Japanese).
25. M.-S. Zhu and C.-X. Lu, *J. Chem. Eng. Data* **39**:205 (1994).