J. Chem. Eng. Data **2003,** 48, 1360−1363

Surface Tension Measurements of Propane (R-290) and Isobutane (R-600a) from (253 to 333) K

Hong Lin and Yuan-Yuan Duan*

Department of Thermal Engineering, Tsinghua University, Beijing 100084, China

---

The surface tensions of propane (R-290) and isobutane (R-600a) were measured over the temperature range (253 to 333) K using a differential capillary rise method (DCRM) under vapor−liquid equilibrium conditions. The temperature and surface tension uncertainties were estimated to be within ±10 mK and ±0.15 $\mathrm{mN \cdot m^{-1}}$, respectively. The present data were used to develop van der Waals-type surface tension correlations for propane (R-290) and isobutane (R-600a). Present results were compared with the available literature data.

Introduction

Because of the ozone layer depletion and global warming caused by chlorofluorocarbon (CFC) and hydrochlorofluorocarbon (HCFC) refrigerants, hydrocarbons (HCs) are expected to be acceptable alternative refrigerants and especially to be a component in mixtures because of their good thermodynamic properties and their universal availability at low price. Surface tension is a basic thermophysical property, related to vapor−liquid interfacial effects that must be known to analyze the heat transfer through heat-exchanging surfaces with bubbles or fluid drops. But up to now, only a few papers have been published on the surface tensions of propane (R-290) and isobutane (R-600a). The surface tension of R-290 was measured by Baidakov and Sulla[1] in the temperature range from (273 to 365) K, by Baydakov[2] from (80 to 288) K, by Stegemeier[3] from (349 to 367) K, and by Vargaftik[4] from (143 to 234) K, respectively. The surface tension of R-600a was measured by Baidakov and Sulla[1] in the temperature range from (273 to 404) K, by Baydakov[2] from (101 to 287) K, by Vargaftik[4] from (173 to 244) K, and by Coffin and Maass[5] from (255 to 334) K, respectively.

This paper presents surface tension measurements for R-290 and R-600a in the temperature range from (253 to 333) K. The experimental results of R-290 and R-600a were represented with van der Waals-type surface tension correlations.

Experimental Section

Materials. The samples of R-290 and R-600a were purchased from Tianjin Chemical Agent Corp., China, and were used without further purification. The manufacturer stated that the purities of R-290 and R-600a were better than 99.9 mass %. A gas chromatograph with a thermal conductivity detector (TCD) and an ODPN column was used to examine the samples, and no impurities were detected.

Apparatus and Procedure. The surface tension was precisely measured using the direct optical measurement system shown in Figure 1 based on the differential capillary rise method. The apparatus has been described in detail[6−10] and is only briefly described here. The measurement system includes a movable coordinate frame, a telescope, and an accurate displacement sensor.

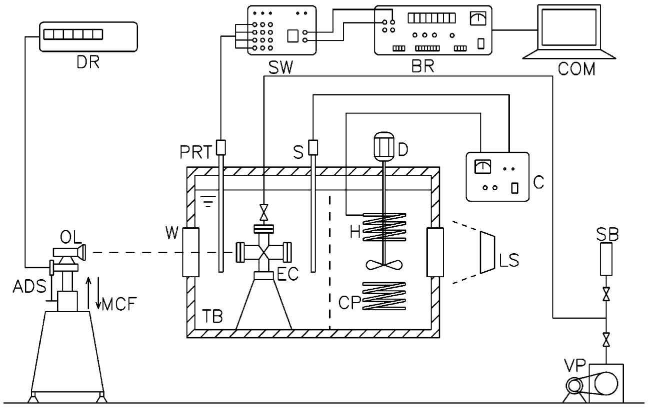

Figure 1. Surface tension apparatus: MCF, movable coordinate frame; OL, optical level instrument; ADS, accurate displacement sensor; DR, digital readout; EC, experimental cell; TB, thermostated bath; H, heater; CP, cooling pipe; D, dynamic−electric stirrer; S, thermal-sensitive resistance sensor; C, controller; PRT, platinum resistance thermometer; SW, selector switch; BR, automatic thermometer bridge; COM, computer; W, window; LS, light source; VP, vacuum pump; SB, sample bottle.

The sample cell contains three capillaries with inner radii of $r_1 = (0.499 \pm 0.001)$ mm, $r_2 = (0.390 \pm 0.001)$ mm, and $r_3 = (0.259 \pm 0.001)$ mm. The three capillary radii were determined by partially filling the capillaries with mercury slugs of different masses.

The sample cell was installed in a thermostated bath. The bath temperature could be varied from (233 to 453) K. The temperature instability in the bath is less than ±5 mK in 8 h. The temperature measurement system includes platinum resistance thermometers (Tinsley: 5187SA) with an uncertainty of ±2 mK, a precision thermometer bridge (Tinsley: 5840D) with an uncertainty within ±1 mK, a select switch (Tinsley: 5840CS/6T), and a personal computer. The overall temperature uncertainty including the bath instability and the temperature measurement system was less than ±10 mK. The differences between the capillary rise heights were determined by measuring the meniscus locations in each capillary through a transparent

---

* Corresponding author. Telephone: +86-10-6278-2484. Fax: +86-10-6277-0209. E-mail address: yyduan@te.tsinghua.edu.cn.

10.1021/je034093m CCC: $25.00

Table 1. Experimental Surface Tension Data and Capillary Constants for R-290

			a^2/mm^2		σ/mN·m^{-1}	
T/K	ρ_l/kg·m^{-3}	ρ_v/kg·m^{-3}	(3, 2)	(3, 1)	(3, 2)	(3, 1)
253.08	554.8	5.5	4.823	4.823	12.98	12.98
254.80	552.7	5.8	4.739	4.737	12.70	12.69
256.89	550.0	6.2	4.646	4.646	12.38	12.38
258.68	547.8	6.6	4.605	4.601	12.21	12.20
260.71	545.2	7.1	4.528	4.539	11.94	11.97
262.70	542.6	7.5	4.410	4.415	11.56	11.58
265.09	539.5	8.1	4.343	4.348	11.31	11.32
267.09	536.9	8.6	4.272	4.269	11.06	11.05
268.88	534.5	9.1	4.197	4.199	10.81	10.81
270.72	532.0	9.6	4.110	4.114	10.52	10.53
273.20	528.7	10.4	4.004	4.004	10.17	10.17
275.38	525.7	11.1	3.953	3.953	9.97	9.97
277.43	522.8	11.7	3.879	3.879	9.71	9.71
279.39	520.1	12.4	3.777	3.777	9.40	9.40
281.48	517.1	13.2	3.703	3.704	9.14	9.15
289.27	505.8	16.4	3.395	3.396	8.14	8.15
293.85	498.9	18.5	3.180	3.180	7.49	7.49
295.73	496.0	19.4	3.097	3.097	7.23	7.23
297.40	493.4	20.3	3.035	3.035	7.04	7.04
299.41	490.2	21.4	2.981	2.990	6.85	6.87
301.45	487.0	22.5	2.907	2.907	6.62	6.62
303.54	483.5	23.8	2.809	2.816	6.33	6.34
305.17	480.8	24.8	2.778	2.782	6.21	6.22
308.01	476.0	26.6	2.661	2.663	5.86	5.86
310.73	471.3	28.5	2.560	2.560	5.55	5.55
314.39	464.9	31.2	2.381	2.380	5.06	5.06
315.71	462.5	32.3	2.301	2.297	4.85	4.84
317.64	458.9	33.9	2.216	2.222	4.62	4.63
319.43	455.6	35.4	2.145	2.148	4.42	4.42
321.41	451.8	37.2	2.094	2.095	4.25	4.26
323.34	448.1	39.0	2.001	2.003	4.01	4.02
325.32	444.1	40.9	1.922	1.927	3.80	3.81
327.40	439.9	43.1	1.851	1.859	3.60	3.62
329.79	434.8	45.7	1.739	1.738	3.32	3.31
331.79	430.6	47.9	1.662	1.667	3.12	3.13
333.30	427.3	49.7	1.589	1.593	2.94	2.95

window of the sample cell using a traveling telescope with an uncertainty of ±10 μm. The differential capillary rise between two capillaries was sensed by a height transducer on the telescope using a digital readout. The contact angle, θ, was assumed to be zero, since the inner walls of the capillaries were carefully washed before the measurements.

All of the measurements were carried out under equilibrium conditions between the liquid and its saturated vapor. A total error analysis gave the estimated surface tension and temperature measurement accuracies to be within ±0.15 mN·m^{-1} and ±10 mK, respectively.

Results and Discussion

The capillary constant is

$$a^2 = 2\sigma/(\rho_l - \rho_v)g \qquad (1)$$

where σ is the surface tension, g is the local gravitational acceleration (at Beijing China g = 9.8015 m·s^{-2}), and ρ_l and ρ_v are the densities of the saturated liquid and vapor, respectively. ρ_l and ρ_v were calculated from available literature data for R-290 and R-600a,[11] and the temperatures have been converted from ITPS-68 to ITS-90.

The height h_0 of the bottom of the meniscus in each capillary was measured, and the actual rise height h was calculated using the Rayleigh correlation.[12] The capillary constant can be represented in relation to the rise heights as

$$a^2 = \frac{h_1 - h_2}{(1/r_1) - (1/r_2)} \qquad (2)$$

Table 2. Experimental Surface Tension Data and Capillary Constants for R-600a

			a^2/mm^2		σ/mN·m^{-1}	
T/K	ρ_l/kg·m^{-3}	ρ_v/kg·m^{-3}	(3, 2)	(3, 1)	(3, 2)	(3, 1)
255.02	600.9	2.2	5.061	5.061	14.85	14.85
256.94	598.8	2.4	5.026	5.026	14.69	14.69
258.83	596.7	2.6	4.932	4.936	14.36	14.37
261.09	594.2	2.8	4.890	4.888	14.17	14.17
262.95	592.1	3.0	4.858	4.859	14.02	14.03
264.86	590.0	3.2	4.765	4.772	13.70	13.72
267.09	587.5	3.5	4.704	4.695	13.46	13.44
268.86	585.5	3.7	4.615	4.615	13.16	13.16
270.90	583.2	4.0	4.564	4.568	12.95	12.97
273.38	580.4	4.3	4.492	4.491	12.68	12.68
275.39	578.1	4.6	4.473	4.484	12.57	12.60
277.16	576.0	4.9	4.391	4.391	12.29	12.29
279.34	573.5	5.2	4.332	4.332	12.06	12.06
281.30	571.2	5.6	4.248	4.240	11.77	11.75
283.29	568.8	5.9	4.214	4.214	11.62	11.62
285.43	566.3	6.3	4.110	4.109	11.28	11.28
287.50	563.8	6.7	4.081	4.078	11.14	11.13
289.31	561.6	7.1	3.995	3.995	10.86	10.86
291.65	558.8	7.6	3.977	3.967	10.74	10.72
293.66	556.3	8.1	3.910	3.910	10.50	10.50
295.75	553.7	8.5	3.848	3.849	10.28	10.28
297.83	551.1	9.1	3.769	3.782	10.01	10.05
299.48	549.1	9.5	3.723	3.723	9.84	9.84
301.43	546.6	10.0	3.639	3.650	9.57	9.60
303.91	543.4	10.7	3.591	3.592	9.37	9.38
305.75	541.0	11.3	3.520	3.520	9.14	9.14
307.41	538.8	11.8	3.479	3.476	8.98	8.98
309.49	536.1	12.4	3.409	3.418	8.75	8.77
311.81	533.0	13.2	3.344	3.342	8.52	8.51
313.49	530.7	13.8	3.272	3.280	8.29	8.31
315.82	527.6	14.6	3.204	3.217	8.05	8.09
317.75	524.9	15.4	3.166	3.170	7.90	7.91
319.32	522.8	16.0	3.098	3.092	7.69	7.68
321.96	519.0	17.1	3.031	3.033	7.45	7.46
323.71	516.5	17.8	2.965	2.956	7.25	7.22
325.37	514.2	18.6	2.892	2.897	7.02	7.04
327.80	510.6	19.7	2.824	2.831	6.79	6.81
329.49	508.2	20.5	2.809	2.809	6.71	6.71
331.68	504.9	21.6	2.710	2.710	6.42	6.42
333.28	502.4	22.5	2.676	2.676	6.29	6.29

Therefore, the surface tension can be calculated from

$$\sigma = \frac{(\rho_l - \rho_v)g}{2}\,\frac{(h_1 - h_2)}{[(1/r_1) - (1/r_2)]} \qquad (3)$$

Using the differential capillary rise method (DCRM) based on the height difference between two different capillaries, two sets of surface tension results were obtained using the height difference between capillaries 3 and 2 and between capillaries 3 and 1. The results are reported in Tables 1 and 2 for R-290 and R-600a, respectively. The saturated liquid density, ρ_l, the saturated vapor density, ρ_v, and the capillary constant, a^2, are also listed in Tables 1 and 2.

The capillary constant a^2 is fitted to the equation

$$a^2 = {a_0}^2\tau^{0.935}(1 + a_1\tau) \qquad (4)$$

where $\tau = 1 - T/T_c$ is the reduced temperature and a_0 and a_1 are the fitting parameters. On the basis of the present results, the values of a_0 and a_1 for R-290 and R-600a have been determined by least-squares fitting and listed in Table 3. The capillary constants, a^2, for R-290 and R-600a versus temperature are plotted in Figure 2.

The surface tension is normally correlated as a function of temperature by a van der Waals-type correlation:

$$\sigma = \sigma_0\left(1 - \frac{T}{T_c}\right)^n \qquad (5)$$

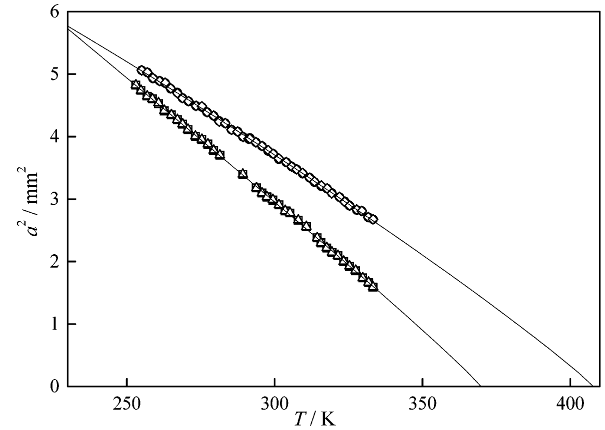

Figure 2. Relation between the temperature and the capillary constant. R-290: □, capillaries 3 and 2; △, capillaries 3 and 1. R-600a: ○, capillaries 3 and 2; ◇, capillaries 3 and 1.

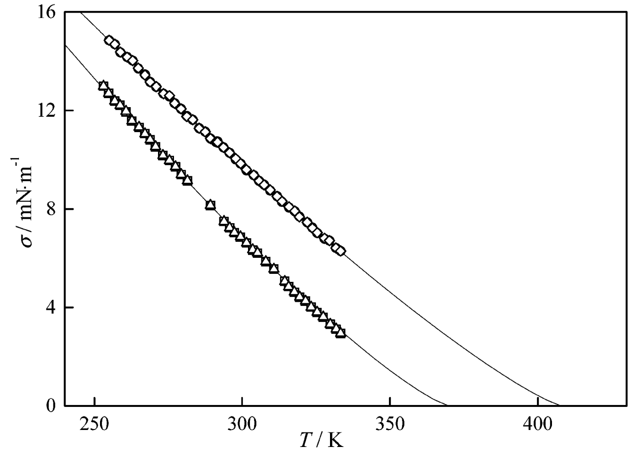

Figure 3. Surface tension vs temperature. R-290: □, capillaries 3 and 2; △, capillaries 3 and 1. R-600a: ○, capillaries 3 and 2; ◇, capillaries 3 and 1.

Table 3. Numerical Constants in Eqs 4 and 5

refrigerants	a_0^2/mm^2	a_1	σ_0/mN·m^{-1}	n	T_c/K
R-290	13.859	0.068	55.817	1.266	369.85[11]
R-600a	13.333	-0.137	48.255	1.201	407.85[11]

where σ_0 and n are empirical constants obtained from a least-squares fit of the experimental data. The value of n usually falls between 1.2 and 1.3 for most fluids. The experimental data in Tables 1 and 2 were used to calculate the values of σ_0 and n for R-290 and R-600a, and the results were listed in Table 3. The average absolute deviations of the present experimental data of R-290 and R-600a from eq 5 are 0.036 mN·m^{-1} and 0.035 mN·m^{-1}, respectively. Figure 3 gives the relations between temperature and surface tension for R-290 and R-600a. Figure 4 shows the absolute deviations of the present data and other available literature data[1−3] from eq 5 for R-290 in the temperature range (220 to 360) K. From Figure 4, it is apparent that the experimental results of Baidakov and Sulla[1] agree well with the present results. Baydakov's data[2] scatter within ±0.1 mN·m^{-1} for temperatures above 253 K, and they show a systematic deviation from eq 5 for temperatures below 253 K. The absolute deviations of Stegemeier's measurements[3] from eq 5 are within ±0.2 mN·m^{-1}, but his data are (0.1−0.2) mN·m^{-1} higher than those of eq 5 when the temperature is above 340 K. Figure 5 shows the absolute deviations of the present data and the experiment data reported by other investigators[1,2,4,5] from eq 5 for R-600a in the temperature range (200 to 407) K. The results in Figure 5 show that most of Baidakov and Sulla's data[1] scatter within ±0.2 mN·m^{-1}. The measurements of Coffin and Maass[5] scatter within ±0.6 mN·m^{-1}. All of the literature data[2,4,5] are systematically higher than those of eq 5 for temperatures below 253 K; this might be caused by a deficiency in the coefficients of eq 5 at low temperatures. The deviation plots also show that the deviations of the present results for R-290 and R-600a are within ±0.15 mN·m^{-1}, which is consistent with the estimated uncertainty. All of the temperatures of the literature data[1−5] have been converted to ITS-90.

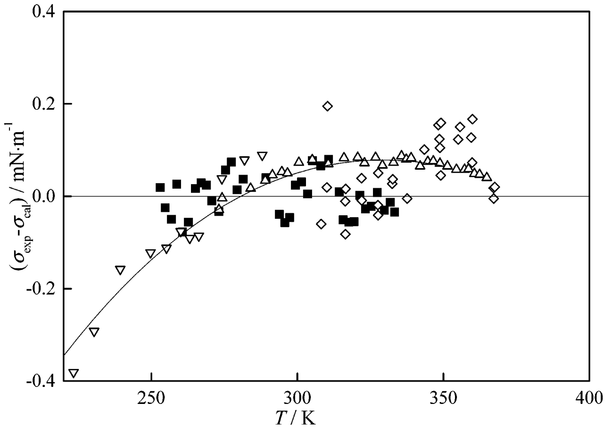

Figure 4. Absolute deviations of experimental surface tension data of R-290 from eq 5: ■, present work; △, Baidakov;[1] ▽, Baydakov;[2] ◇, Stegemeier;[3] —, Baidakov.[1]

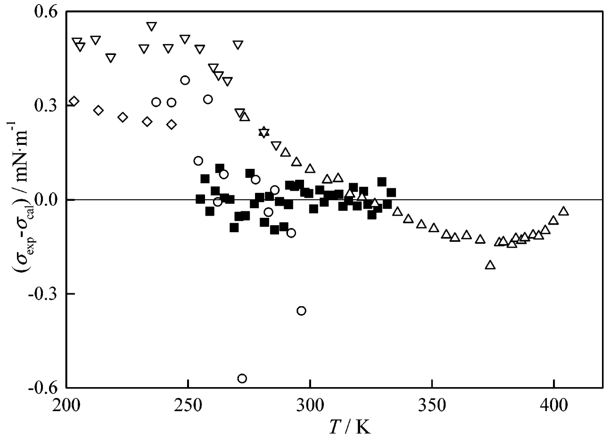

Figure 5. Absolute deviations of experimental surface tension data of R-600a from eq 5: ■, present work; △, Baidakov;[1] ▽, Baydakov;[2] ◇, Vargaftik;[4] ○, Coffin.[5]

Conclusions

The surface tensions of R-290 and R-600a were measured over a wide temperature range using the differential capillary rise method. The uncertainty of the surface tension measurements was estimated to be within ±0.15 mN·m^{-1}. The experimental data were then used to develop correlations for the surface tensions of R-290 and R-600a.

Literature Cited

(1) Baidakov, V. G.; Sulla, I. I. Surface Tension of Propane and Isobutane at Near-critical Temperatures. *Russ. J. Phys. Chem.* **1985**, *59*, 551−552.
(2) Baydakov, V. G. The Capillary Constant and Surface Tension of Low-temperature Propane and Isobutane. *Fluid Mech.—Sov. Res.* **1985**, *14*, 118−122.
(3) Stegemeier, G. L. Interfacial Tension of Synthetic Condensate Systems. Ph.D. Thesis, University of Texas, 1959.

(4) Vargaftik, N. B. *Tables on the Thermophysical Properties of Liquids and Gases: In Normal and Dissociated State*; Hemisphere: Washington, DC, 1975.

(5) Coffin, C. C.; Maass, O. The Preparation and Physical Properties of α-, β- and γ-butane and Normal and Isobutane. *J. Am. Chem. Soc.* **1928**, *50*, 1427−1437.

(6) Zhu, M. S.; Han, L. Z.; Lu, C. X. Surface Tension of HFC-134a. *Fluid Phase Equilib.* **1993**, *86*, 363−367.

(7) Zhu, M. S.; Lu, C. X. Surface Tension of Difluoromethane. *J. Chem. Eng. Data* **1994**, *39*, 11−12.

(8) Duan, Y. Y.; Shi, L.; Zhu, M. S.; Han, L. Z. Surface Tension of trifluoroiodomethane (CF_3I). *Fluid Phase Equilib.* **1999**, *154*, 71−77.

(9) Duan, Y. Y.; Shi, L.; Zhu, M. S.; Han, L. Z.; Lei, X. Surface Tension of Pentafluoroethane and 1,1,1,2,3,3,3-Heptafluoropropane. *Fluid Phase Equilib.* **2000**, *172*, 237−244.

(10) Duan, Y. Y.; Lin, H.; Wang, Z. W. Surface Tension Measurements of Difluoromethane (R-32) and the Binary Mixture Difluoromethane (R-32) + 1,1,1,2-tetrafluoroethane (R-134a) from (253 to 333) K. *J. Chem. Eng. Data* **2003**, *48*, 1068−1072.

(11) Younglove, B. A.; Ely, J. F. Thermophysical Properties of Fluids. II. Methane, Ethane, Propane, Isobutane and Normal Butane. *J. Phys. Chem. Ref. Data* **1987**, *16*, 577−798.

(12) Rayleigh, O. M. On the Theory of Capillary Tube. *Proc. R. Soc. London, Ser. A* **1916**, *92*, 184−195.

Received for review May 16, 2003. Accepted July 3, 2003. This work was supported by the National Natural Science Foundation of China (No. 50225622) and SRFDP (No. 20010003015).

JE034093M