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Surface Tension of Normal Pentane, Hexane, Heptane, and Octane

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New experimental results are reported for the surface tension of normal pentane, hexane, heptane, and octane. The data were obtained with a differential capillary-rise method and cover the entire liquid range from the triple-point temperature to the critical temperature. Equations for calculating the surface tension of these *n*-alkanes as a function of temperature are presented.

KEY WORDS: alkanes; capillary length; critical temperature; heptane; hexane; hydrocarbons; octane; pentane; surface tension.

1. INTRODUCTION

This paper is concerned with the surface tension of some technically important hydrocarbons, namely, *n*-pentane (C_5H_{12}), *n*-hexane (C_6H_{14}), *n*-heptane (C_7H_{16}), and *n*-octane (C_8H_{18}). The surface-tension data reported in the literature for these fluids do not cover the entire temperature range of the liquid state [1–11]. Specifically, sufficient information for the surface tension is lacking at lower temperatures and in the temperature range near the critical temperature. We have therefore made an experimental study of the surface tension of these normal alkanes at temperatures from the triple-point temperature to the critical temperature. Representative equations representing the surface tension of these *n*-alkanes as a function of temperature are also presented.

2. EXPERIMENT

In our experiments we have used a differential capillary-rise method similar to that used by Gielen et al. [12] for other fluids. A schematic

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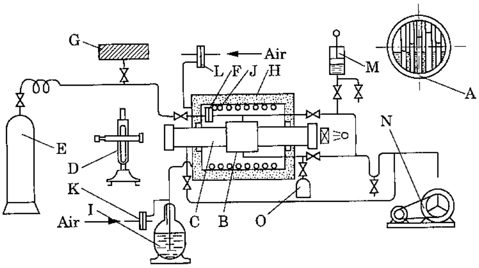

Fig. 1. Schematic illustration of the experimental arrangement.

representation of the experimental arrangement is shown in Fig. 1. An assembly of five round glass capillaries located vertically in a circular container (A) is placed in the measurement cell (B). The measurement cell (B) has two quartz windows with two cylindrical observation tubes (C). The windows are sealed with Teflon O-rings for the experiments at temperatures below 293 K and with silver O-rings for the experiments at temperatures above 293 K. The observation tubes (C) are evacuated to prevent condensation of water on the windows. Prior to the assembly of the apparatus the inner radii of the five selected capillaries were determined by measuring the weight of threads of mercury in the capillaries [13]. The radii are $r_1 = 0.4990$ mm, $r_2 = 0.1752$ mm, $r_3 = 0.1582$ mm, $r_4 = 0.1156$ mm, and $r_5 = 0.3390$ mm, measured with a maximum uncertainty of 0.05%. The vertical distance Δh_{pq} between the menisci in two capillaries, p and q, is measured with the aid of a cathetometer (D) with an accuracy of about ± 0.005 mm.

The vapor pressure of the liquid is balanced against argon pressure from a supply cylinder (E) with the aid of the membrane transducer (F). The argon pressure is measured with a digital pressure gauge (G) with an accuracy varying from about 5 to 8 mbar. The measurement cell (B), as well as the pressure transducer (F), is located inside the liquid thermostat (H). At temperatures below 293 K, methanol is used as the thermostat fluid, while silicon oil PES-2 is used at temperatures above 293 K. At low temperatures the thermostat is cooled with liquid nitrogen from a Dewar vessel (I) and at high temperatures the thermostat is heated by an electrical heater (J). The flow rate of nitrogen is controlled with the aid of valves (K and L). The temperature is maintained with an automatic controller at a desired temperature to within ± 0.015 K. The temperatures are measured with standard platinum resistance thermometers (a TSPN–Pt resistance thermometer at low temperatures and a PTS–10 Pt resistance thermometer at the higher temperatures), calibrated at the All-Union Research Institute of Metrology and Standards in Moscow against the International Practical Temperature Scale of 1968. The temperatures were measured with a precision of about 1 mK and with an absolute accuracy of 0.02 K.

The hydrocarbons were supplied by the Grosneftehim Company in Grozny with a stated purity of 99.90 mol% for C_5H_{12} and C_6H_{14} and of 99.89 mol% for C_7H_{16} and C_8H_{18}. All parts of the experimental apparatus in contact with the measurement fluid were carefully cleaned initially. After the apparatus had been assembled the measurement cell was rinsed with a liquid introduced into the cell from an additional supply (M). Subsequently, the cell was evacuated with the aid of vacuum pump (N). Then a prescribed amount of the sample fluid from the supply vessel (O) was put in the cell to obtain a density within 0.5% from the critical density. A more detailed discussion of the experimental apparatus and method has been presented elsewhere [14, 15].

3. RESULTS

From the experimental measurements of the differential capillary rise Δh_{pq}, a corresponding value, a^2_{pq}, for the squared capillary-length parameter a^2, also called the Sugden parameter, was calculated with an iteration method described by Sugden [16]. The final experimental value of a^2 was then taken as the average $\langle a^2_{\mathrm{pq}} \rangle$ of all the individual data obtained at a given temperature. The results thus obtained for a^2 as a function of temperature are presented in Table I and plotted in Fig. 2 as a function of temperature.

Special care was taken to study the behavior of the meniscus in the vicinity of the critical temperature. Here the temperature was varied slowly at a rate of a few tenths of a degree per hour and the menisci were observed at temperature intervals of 0.005 K. The behavior of the interface near the critical temperature, including color effects, was similar to that described by Lakoza and Chalyi [17]. The critical temperature T_c was determined as the average of the temperatures of appearance and disappearance of the meniscus, which differed by less than about 0.03 K. The values thus obtained for T_c are presented in Table II, which also includes values of T_c reported by other investigators. The values observed for T_c are known to be very sensitive to the presence of impurities.

The surface tension σ is related to the capillary constant a^2 by [23]

$$\sigma = \frac{1}{2\cos\theta} a^2 g \, \Delta\rho \tag{1}$$

where θ is the contact angle, g the gravitational acceleration constant, and $\Delta\rho$ the difference between the densities of the coexisting liquid and vapor

Table I. Experimental Results for Normal Pentane, Hexane, Heptane, and Octane

T (K)	$\Delta\rho$ (kg · m^{-3})	a^2 (mm^2)	σ (mN · m^{-1})	T (K)	$\Delta\rho$ (kg · m^{-3})	a^2 (mm^2)	σ (mN · m^{-1})
			n-Pentane				
144.18	742.8	9.247	33.76	407.25	433.8	1.963	4.17
145.08	742.7	9.214	33.54	438.91	301.2	1.011	1.49
145.23	742.6	9.210	33.53	455.09	202.2	0.502	0.50
156.53	742.0	8.909	32.40	459.98	167.1	0.342	0.28
165.87	738.1	8.659	31.32	467.28	109.9	0.092	0.05
181.01	727.2	8.254	29.42	467.69	106.5	0.078	0.04
193.45	715.2	7.920	27.77	468.02	103.8	0.065	0.03
226.09	677.6	7.039	23.38	468.53	99.5	0.046	0.02
249.34	650.1	6.406	20.42	469.15	94.3	0.022	0.01
270.21	627.3	5.836	17.94	469.28	93.2	0.017	0.008
292.49	605.3	5.222	15.49	469.51	91.2	0.0077	0.0034
336.52	563.7	3.995	11.04	469.67	89.9	0.0005	0.002
371.17	518.3	3.010	7.65				
			n-Hexane				
175.12	756.6	8.559	31.74	402.71	535.5	2.911	7.64
178.03	755.1	8.489	31.42	447.13	442.8	1.738	3.77
181.52	753.2	8.405	31.03	448.93	326.6	0.778	1.25
189.32	748.5	8.217	30.15	497.64	256.3	0.318	0.40
198.74	742.0	7.990	29.06	504.32	222.3	0.109	0.119
213.54	730.4	7.632	27.32	505.11	218.2	0.083	0.089
244.81	702.5	6.870	23.66	506.31	211.7	0.042	0.044
270.93	677.9	6.230	20.70	506.48	210.8	0.036	0.037
284.72	664.9	5.890	19.20	506.74	209.4	0.027	0.027
312.58	638.8	5.199	16.28	507.12	207.3	0.013	0.013
352.49	599.7	4.197	12.34	507.31	206.3	0.005	0.005
387.03	558.6	3.316	9.08	507.39	205.9	0.002	0.002
			n-Heptane				
183.21	772.8	8.523	32.28	459.18	488.3	2.125	5.09
187.49	771.1	8.427	31.85	490.41	405.8	1.346	2.68
193.41	768.1	8.294	31.23	497.06	383.2	1.177	2.21
199.93	764.2	8.148	30.52	507.94	341.6	0.896	1.50
207.84	758.6	7.971	29.64	522.17	277.4	0.519	0.705
224.58	744.5	7.594	27.71	532.19	224.8	0.241	0.266
258.14	710.8	6.836	23.82	537.27	195.6	0.092	0.089
278.59	689.3	6.370	21.53	539.02	185.1	0.038	0.034
318.07	650.1	5.466	17.42	539.13	184.4	0.034	0.031
353.49	618.4	4.645	14.08	539.74	180.7	0.014	0.012
385.91	589.5	3.885	11.23	540.08	178.6	0.0013	0.0011
408.33	566.6	3.354	9.31				

Table I. (*Continued*)

T (K)	$\Delta\rho$ ($\mathrm{kg \cdot m^{-3}}$)	a^2 ($\mathrm{mm^2}$)	σ ($\mathrm{mN \cdot m^{-1}}$)	T (K)	$\Delta\rho$ ($\mathrm{kg \cdot m^{-3}}$)	a^2 ($\mathrm{mm^2}$)	σ ($\mathrm{mN \cdot m^{-1}}$)
			n-Octane				
218.51	756.7	7.890	29.26	488.54	487.0	1.972	4.71
224.58	756.7	7.761	28.79	501.27	451.7	1.676	3.71
243.18	750.3	7.366	27.09	522.48	373.7	0.176	2.15
261.92	736.6	6.966	25.15	564.92	117.1	0.114	0.066
279.19	720.1	6.597	23.28	566.74	102.3	0.063	0.032
279.52	719.7	6.590	23.25	567.14	99.0	0.052	0.025
325.37	670.7	5.602	18.42	568.08	91.2	0.024	0.011
354.31	642.6	4.973	15.66	568.56	87.2	0.009	0.004
392.04	611.3	4.145	12.42	568.74	85.6	0.003	0.0012
428.51	580.7	3.319	10.01	568.78	85.3	0.002	0.0006
461.92	539.8	2.582	6.83	568.79	85.2	0.001	0.0005

phases. For the fluids considered here, the liquid wets the glass capillary and the contact angle θ was considered to be zero. In order to convert the experimental results for a^2 into values for the surface tension, we calculated densities for the saturated vapor and liquid phases from data previously measured in the Grozny Petroleum Institute [24–29] and for *n*-pentane and *n*-heptane also from information available elsewhere in the literature [30]. The values thus obtained for the density difference $\Delta\rho$ and the resulting values for the surface tension σ are also included in Table I. The surface tension is plotted as a function of temperature in Fig. 3.

Table II. Critical Temperatures T_c (K) of Normal Pentane, Hexane, Heptane, and Octane

C_5H_{12}	C_6H_{14}	C_7H_{16}	C_8H_{18}
469.68[a]	507.43[a]	540.11[a]	568.82[a]
469.60[b]	507.35[b]	540.2[b]	568.72[b]
469.62[c]	507.25[e]	539.86[e]	
470.4[d]	507.55[f]	540.8[f]	

[a] This work.
[b] Source: Ref. 18.
[c] Source: Ref. 19.
[d] Source: Ref. 20.
[e] Source: Ref. 21.
[f] Source: Ref. 22.

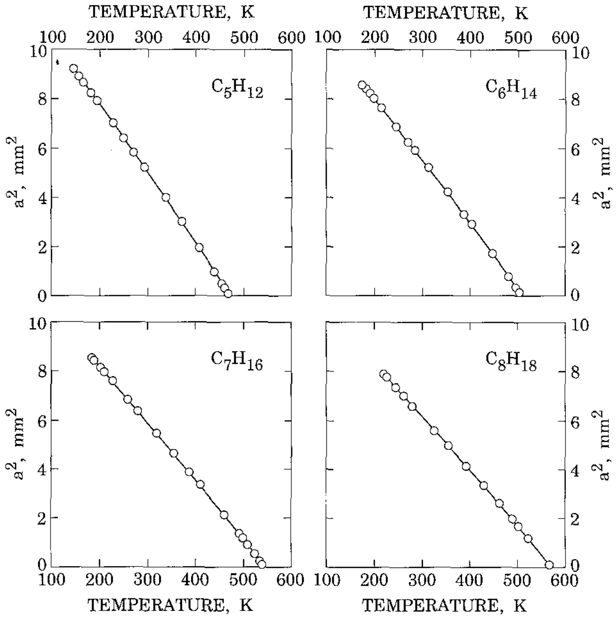

Fig. 2. The squared capillary-length parameter a^2 as a function of temperature. The circles indicate the experimental data and the curves represent the values calculated from Eq. (10).

In the temperature range of overlap, our data for the surface tension of *n*-pentane agree with those reported by Rossini et al. [1] and by Jasper et al. [2] within 0.8%, for *n*-hexane with the surface tensions reported by Pugachevich and Cherkasskaj [10] within 0.5%, for *n*-heptane with the values reported by Jasper et al. [2], Benikovsky et al. [6], and Pugachevich and Khvorov [11] within 0.5%, and for *n*-heptane and *n*-octane with the data of Volyak [4, 5] within 1.5%. The experimental values obtained by Muratov et al. [9] for *n*-heptane at temperatures near the critical temperature agree with our results within about 2%.

4. DISCUSSION

Near the critical temperature the surface tension σ and the capillary-length parameter a^2 satisfy power laws of the form [23]

$$\sigma = \sigma_0 \tau^{\mu} \tag{2}$$

$$a^2 = a_0^2 \tau^{\phi} \tag{3}$$

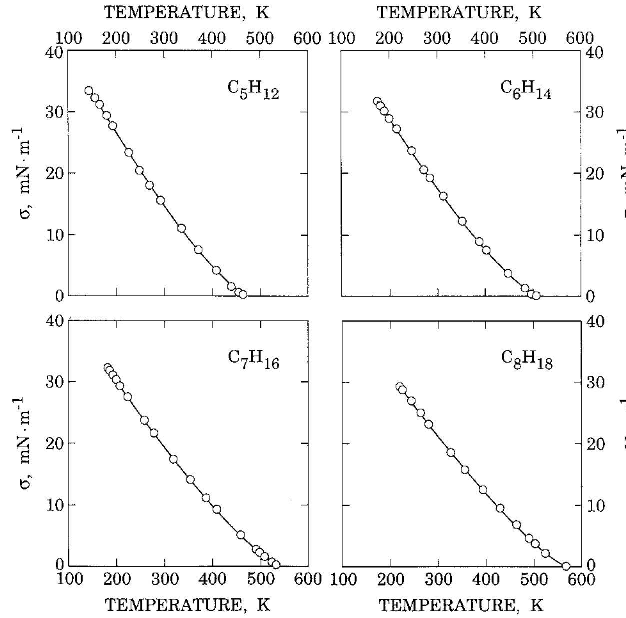

Fig. 3. The surface tension σ as a function of temperature. The circles indicate the experimental data and the curves represent the values calculated from Eq. (13).

where τ is the reduced temperature difference

$$\tau = 1 - T/T_c \tag{4}$$

Furthermore, the difference $\Delta\rho$ between the liquid and the vapor density below the critical temperature and the specific heat c_v at the critical isochore $\rho = \rho_c$ above the critical temperature satisfy asymptotic critical power laws of the form [31]

$$\Delta\rho = 2\rho_c B\tau^{\beta} \tag{5}$$

$$c_v = T_c^{-1}\rho^{-1}P_c C_0 \,|\tau|^{-\alpha} \tag{6}$$

where ρ_c is the critical density and P_c the critical pressure. The exponents μ, ϕ, β, and α are universal; for fluids they have the same values as those for the three-dimensional Ising model [31, 32], while σ_0, a_0^2, B, and C_0 are system-dependent amplitudes. The asymptotic behavior Eq. (2) of the surface tension is related to the asymptotic behavior Eq. (6) of the specific heat by the so-called principle of two-scale-factor universality [12, 33, 34].

Specifically, as done by Chaar et al. [35], it is convenient to introduce a quantity Y such that

$$Y=\sigma[\alpha\tau^2\rho_c c_v/k_B]^{-2/3}/k_B T_c \tag{7}$$

where k_B is Boltzmann's constant. Asymptotically close to the critical temperature this quantity Y has a universal value which is the same for all fluids and which is currently estimated as [35, 36]

$$Y=5.73\pm 0.16 \tag{8}$$

From the relations above it follows that $3\mu=4-2\alpha$ and $\phi=\mu-\beta$. Here we adopt the values $\alpha=0.11$ and $\beta=0.325$ in agreement with the theoretical predictions [31, 32], so that

$$\mu=1.26, \qquad \phi=0.935 \tag{9}$$

The theoretical validity of the simple critical power laws is restricted to a small range of temperatures near the critical temperature [37]. However, it has been noted that the power law (3) for a^2 is applicable over an appreciably larger temperature range [34, 38, 39]. The theoretical reason for this observation is unclear, although it may be noted that the actual value of the exponent ϕ is close to its classical value of unity [34]. Even so, a simple power law cannot be expected to be applicable in the entire liquid temperature range covered by our measurements. Nevertheless, it is found that a simple equation of the form

$$a^2=a_0^2\tau^{0.935}[1+a_1\tau^{1/2}] \tag{10}$$

containing one correction-to-scaling term [31], yields a surprisingly good representation of a^2 at temperatures all the way down to the triple-point temperature. The values of the coefficients a_0^2 and a_1 deduced from least-squares fits to the experimental data are given in Table III. Corresponding

Table III. Coefficients in Eqs. (10) and (13) for a^2 and σ

	T_c (K)	a_0^2 (mm^2)	a_1	C_0	σ_0[a] (mN·m^{-1})	σ_1[b]	SD[c] (mN·m^{-1})
C_5H_{12}	469.68	12.916	+0.0094	30.76	53.94	−0.0048	0.18
C_6H_{14}	507.43	12.753	−0.0039	33.55	54.78	−0.0117	0.07
C_7H_{16}	540.11	12.520	+0.0035	36.75	55.49	−0.0253	0.10
C_8H_{18}	568.82	12.217	+0.0208	38.60	54.77	−0.0114	0.14

[a] Calculated from specific-heat amplitude C_0.

[b] From fit to experimental surface-tension data.

[c] Standard deviation of fit to surface-tension data.

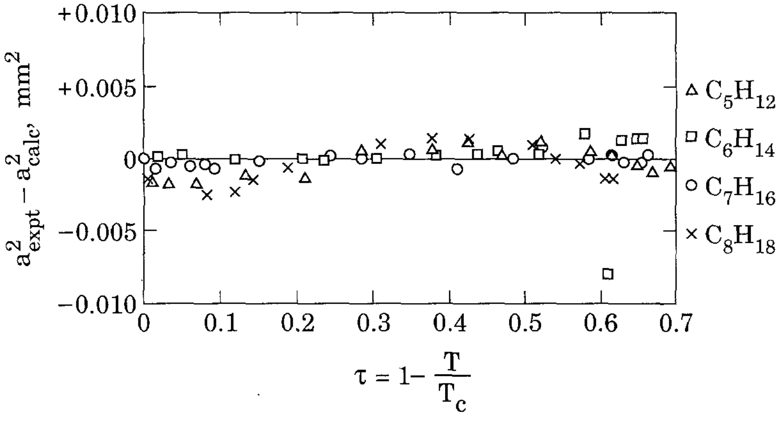

Fig. 4. Differences $a^2_{expt} - a^2_{calc}$ between the experimental values of a^2 and the values calculated from Eq. (10).

deviation plots are shown in Fig. 4. With few exceptions Eq. (10) reproduces most experimental data for a^2 within 0.003 mm^2 as can be seen from Fig. 4.

Substitution of the power laws given by Eqs. (2) and (6) into the two-scale-factor relation (7) yields [35]

$$Y = \sigma_0(\alpha P_c C_0)^{-2/3} (k_B T_c)^{1/3} \tag{11}$$

Equations (8) and (11) relate the amplitude σ_0 of the power law for the surface tension to the amplitude of the power law for the specific heat and, hence, to derivatives of the equation of state. Recently, Kurumov and Grigoryev have developed a unified scaled equation of state for the normal alkanes in the critical region [40]. In this unified equation of state the system-dependent constants are represented as polynomials in terms of the Pitzer acentric factor ω. The specific-heat amplitude $C_0(\omega)$ is related to the coefficients $a(\omega)$ and $k(\omega)$ in the unified scaled equation of state by [41]

$$C_0 = ak\frac{\gamma^2(1-\gamma)(\gamma+\alpha-1)}{2\alpha(2\gamma+\alpha-2)} \tag{12}$$

with $\gamma = 2 - \alpha - 2\beta = 1.24$. The values thus obtained for C_0 and then for σ_0 through Eq. (11) are included in Table III.

The power law given by Eq. (5) for the density difference is known to be valid very close to the critical temperature only, and the theoretically predicted temperature difference of $\Delta\rho$ is quite complicated [37, 42]. In

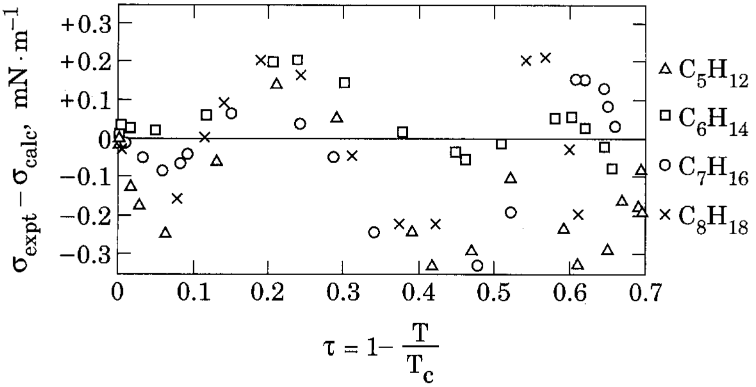

Fig. 5. Differences $\sigma_{expt} - \sigma_{calc}$ between the experimental values of σ and the values calculated from Eq. (13).

analogy to Eq. (10) we have fitted in practice the experimental surface-tension data to

$$\sigma = \sigma_0 \tau^{1.26}[1 + \sigma_1 \tau^{0.5}] \tag{13}$$

treating σ_1 as an adjustable parameter. The values thus obtained for σ_1, together with the standard deviation of the fits to the surface-tension data, are included in Table III. We do not find any significant improvement of the fit when we use a three-term equation like $\sigma = \sigma_0 \tau^{1.26}[1 + \sigma_1 \tau^{0.5} + \sigma_2 \tau]$ or $\sigma = \sigma_0 \tau^{1.26}[1 + \sigma'_1 \tau + \sigma'_2 \tau^2]$ as adopted by Somayajula [43]. Deviation plots for the surface tension σ when calculated from Eq. (13) are shown in Fig. 5. As can be seen from this figure, Eq. (13) represents the surface tension to within about ±0.3 mN · m^{-1}.

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