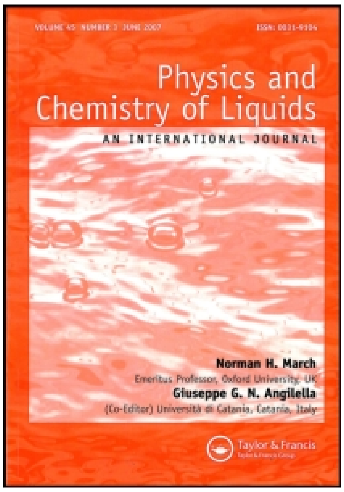

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# Dependence of Surface Tension of Near-boiling Non-associated Liquids on Their Molar Volume and Some Critical Constants

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# DEPENDENCE OF SURFACE TENSION OF NEAR-BOILING NON-ASSOCIATED LIQUIDS ON THEIR MOLAR VOLUME AND SOME CRITICAL CONSTANTS

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The equation proposed for near-boiling non-associated liquids describes a new functional dependence of their surface tension on such physico-chemical characteristics as: critical volume, critical temperature and molar volume at the temperatures which are near their normal boiling points. It is shown that, in the case of some low-boiling liquids, possessing small molecules, this equation can be used for the adequate calculation of surface tension at different temperatures in the liquid phase.

*Keywords*: Surface tension; Non-associated liquids; Molar volume; Critical constants

## 1. INTRODUCTION

Various equations proposed in order to describe surface tension $\sigma$ of non-associated liquids on the basis of their different physico-chemical parameters may be conditionally divided into two parts. One part of equations is based on the usage of parachor which is an empirical additive molecular function of atoms, atomic groups and bonds which form molecular structure (see [1]). Because of this circumstance all these equations have an empirical character and their calculation errors are around 3–4%.

Another part comes mainly from Eqs. (1) and (2):

$$\sigma = \chi(T_c - T)/V^{2/3} \qquad (1)$$

$$\sigma = \alpha T_c^{1/3} P_c^{2/3} (1 - T/T_c)^{\gamma}. \qquad (2)$$

Equation (1) was proposed by Eötvös [2]. It connects the value of $\sigma$ at absolute temperature $T$ with molar volume of a liquid $V$ and its critical temperature $T_c$; $\chi$ is so-called Eötvös constant which is equal to 2.12 for many nonpolar (or low-polar) liquids [1]. Equation (2) was proposed by van der Waals in the work [3]. In this equation $P_c$ is critical pressure of a liquid and $\alpha$ and $\gamma$ are some coefficients which are constant for different liquids. Equations (1) and (2) hold for nonpolar (or low-polar) liquids [1] and their calculation errors are around 3–5%. Here it is to be noted that these equations are not applicable for some nonpolar low-boiling liquids. The presence of such facts caused new values of $\chi$, $\alpha$ and $\gamma$ to be proposed. Various modifications of Eqs. (1) and (2) (see, for instance, the work [4]) were based on the attempt to describe the above-mentioned constants by means of other molecular physico-chemical parameters and, therefore, to extend the calculation possibilities of these equations. However, these new modifications met their limitations [1, 5].

Nevertheless, the existence of Eqs. (1) and (2) demonstrates the importance of molar volume and critical constants for the adequate description of $\sigma$ at different absolute temperatures. The aim of the present work consists in an attempt to find the equation which connects $\sigma$ with these physico-chemical quantities of non-associated liquids.

## 2. THEORY

First of all we shall consider the liquid under investigation in the temperature range which is near its normal boiling point. At this condition all the molecules on its surface may be considered as spherical particles due to their fast mutual reorientations. These surface molecules (hard spheres) take part in vibrational and translational motions. Their spatial translations are realized in the form of irregular jumpings and may lead to their move into the volume of this liquid or into the gas phase (their vaporization). In general case the energy of these jumping molecules $E_j$ is essentially higher than the energy of the vibrating ones. It means that $E_j$ is much more than $kT$

(where $k$ is Boltzmann constant). In consequence of this fact the number of the jumping molecules is essentially less than the number of the vibrating ones. The above mentioned vaporization of the jumping molecules leads to the further decrease in their number on the considered surface. Thus, the surface tension of the investigated liquid will be mainly caused by those surface molecules which vibrate on its surface and, therefore, don't abandon it for a long time. Bearing in mind all the above mentioned reasons, we may take into account only the vibrational motion of the surface molecules. Further we shall consider that these molecular vibrations are harmonic and have their frequency $\nu$ which is the same for any vibrational degree of freedom.

Let the number of the surface molecules of the investigated liquid be $N$. At the above mentioned high temperatures (which depend on the nature of this liquid but, however, are much less than $E_j/k$) we may write (see [6]) the following expression for the partition function $F$ taken over all the vibrational states of these molecules considered as three-dimensional oscillators, possessing the limited number of their vibrational quanta which is defined by $E_j$:

$$F = g^N (kT/\mathrm{h}\nu)^{3N}, \tag{3}$$

where $g$ is factor of electronic state degeneration and h is Planck constant. In the case of the one separate molecule of the liquid, this factor describes the degree of its electronic state degeneration. The free energy $A$ of the considered system of $N$ particles is defined (see [6]) by means of the following general equation of statistical thermodynamics:

$$A = -kT \ln F. \tag{4}$$

Using Eqs. (3) and (4), we shall have:

$$A = -NkT \ln[g(kT/\mathrm{h}\nu)^3] = NkT \ln[g^{-1}(\mathrm{h}\nu/kT)^3]. \tag{5}$$

Let, further, $N$ be equal to $n$. $n$ is the number of the molecules of the mentioned surface which are situated on its unit area. In this case $A$ will be equal to $\sigma$:

$$\sigma = nkT \ln[g^{-1}(\mathrm{h}\nu/kT)^3]. \tag{6}$$

Now we have to define $n$. Bearing in mind this purpose, we shall use the following equality:

$$4\pi r^3/3 = V/N_A, \tag{7}$$

where $r$ is molecular radius, $V$ is molar volume of a liquid and $N_A$ is Avogadro's number. From Eq. (7) we have:

$$r = (3V/4\pi N_A)^{1/3}. \tag{8}$$

Here $r$ is the average value of the effective molecular radius which is calculated for all the molecules of the investigated liquid phase (including its inner molecules). Taking into account that the relative share of the jumping molecules on the surface of the liquid under consideration is less than in the middle of its volume, we can write for the effective radius $r_s$ of the surface vibrating molecules the following equation:

$$r_s = \varphi(3V/4\pi N_A)^{1/3}, \tag{9}$$

where $\varphi$ is coefficient of proportionality. Bearing in mind that the liquid phase is near its boiling point and based on the principle of corresponding states, we shall suppose that $\varphi$ is constant for various non-associated liquids.

Let the surface area occupied by the one separate surface molecule be $s$. Then $n$ will be defined on the basis of the following equalities:

$$s = \pi r^2 = \pi\varphi^2(3V/4\pi N_A)^{1/3} \tag{10}$$

$$n = s^{-1} = \pi^{-1}\varphi^{-2}(4\pi N_A/3V)^{2/3}. \tag{11}$$

Uniting Eqs. (6) and (11), we have:

$$\sigma = \pi^{-1}\varphi^{-2}kT(4\pi N_A/3V)^{2/3}\ln[g^{-1}(\mathrm{h}\nu/kT)^3]. \tag{12}$$

Further, the frequency $\nu$ in Eq. (12) may be received by means of Eq. (13) which holds for every vibrational degree of freedom of the surface molecule considered as a three-dimensional oscillator:

$$E = 2(\pi\nu a)^2 m = kT, \tag{13}$$

where $E$ is the average energy of molecular vibrational motion along one of space axes (see [6]), $a$ is its amplitude and $m$ is molecular mass. In the present work we shall consider that $a$ is proportional to $r_s$:

$$a = fr_s, \tag{14}$$

where $f$ is coefficient of proportionality which doesn't depend on the nature of a non-associated liquid. Combining Eqs. (9), (13) and (14), we have for $\nu$:

$$\nu = (\pi f \varphi)^{-1}(4\pi N_A/3V)^{1/3}(kT/2m)^{1/2}. \tag{15}$$

From Eqs. (12) and (15) it follows that:

$$\sigma = \pi^{-1}\varphi^{-2}kT(4\pi N_A/3V)^{2/3}\ln\{4N_A \mathrm{h}^3[3\pi^{1/2}(f\varphi)^3 gV(2\pi mkT)^{3/2}]^{-1}\}. \tag{16}$$

The quantity $g$ in Eq. (16) may be defined on the basis of the following reasons. Let us consider the investigated liquid at its critical conditions. We shall suppose that the value of $g$ which is defined only by molecular electronic structure (see above) remains constant at different temperatures of liquid range. Further, for any liquid considered at its critical temperature $T_c$ its molecular motion has mainly a translational character [1, 5, 6]. In the case of its one separate molecule, this translational motion is realized in some space cell which has its volume $v$. The value of $v$ can be defined as:

$$v = V_c/N_A, \tag{17}$$

where $V_c$ is molar critical volume of a liquid.

Let $N$ be (as above) the number of the molecules of this liquid. Considering its volume as a cell space (the number of these cells is also equal to $N$), we may write (see [6]) the following expression for the partition function $F$ taken over all the molecular states connected with the translational motion of these molecules:

$$F = g^N[(2\pi mkT_c)^{3/2}ve/\mathrm{h}^3]^N, \tag{18}$$

where $e$ is Napierian base. The presence of $e$ in Eq. (18) indicates that these molecules may have their cell exchange [6]. Combining Eqs. (4), (17) and (18) we shall receive for the free energy $A$ of the investigated

$N$-molecular system:

$$A = NkT\ln\{N_A\mathrm{h}^3[egV_c(2\pi mkT_c)^{3/2}]^{-1}\}. \tag{19}$$

However, the values of $\sigma$ and $A$ go to zero at critical conditions. In this case, it follows from Eq. (19) that:

$$g = N_A\mathrm{h}^3[eV_c(2\pi mkT_c)^{3/2}]^{-1}. \tag{20}$$

Using Eq. (20) in Eq. (16) we have:

$$\sigma = \pi^{-1}\varphi^{-2}kT(4\pi N_A/3V)^{2/3}\ln\{4e[3\pi^{1/2}(f\varphi)^3]^{-1}(V_c/V)(T_c/T)^{3/2}\}. \tag{21}$$

Uniting all the constants of Eq. (21), we shall receive for $\sigma$ the following final equation:

$$\sigma = \xi TV^{-2/3}\ln[\mu(V_c/V)(T_c/T)^{3/2}], \tag{22}$$

where $\xi$ and $\mu$ are constant for any non-associated liquid.

## 3. RESULTS AND DISCUSSION

The practical usage of Eq. (22) requires the numerical values of $\xi$ and $\mu$. In order to appreciate these values the fifteen liquids presented in Table I were investigated. The values of their quantities $V$ and $\sigma$ at their normal boiling points $T$ and also their critical constants $V_c$ and $T_c$ were taken from [5, 7–17]. Here it is to note that the compounds presented in Table I were chosen because of the following reasons: firstly, they are well studied experimentally; secondly, they have various chemical structures which differ significantly. A part of them belongs to inorganic compounds; another one is presented by organic substances. Nevertheless, the above mentioned physico-chemical parameters of these compounds obey Eq. (22). The numerical values of $\xi$ and $\mu$ defined by means of the least-square method on the basis of the data set of Table I are $1.324 \times 10^{-7}$ ($N \times \mathrm{K}^{-1} \times \mathrm{mol}^{-2/3}$) and 0.483 respectively.

The results of the usage of Eq. (22) with the found values of $\xi$ and $\mu$ are presented in Table II. All the necessary critical constants of the investigated substances side by side with the values of $\sigma$ and $V$ at

TABLE I To the definition of quantities of $\xi$ and $\mu$

| | | $T(K)$ | | $V \times 10^6$ ($m^3 \times mol^{-1}$) | | $\sigma \times 10^3$ | ($\sigma V^{2/3} T^{-1} \times 10^7$) | |
|---|---|---|---|---|---|---|---|---|
| *Ns.* | *Compound:* | *Boil.* | *Crit.* | *Boil.* | *Crit.* | ($N \times m^{-1}$) | ($J \times K^{-1} \times mol^{-2/3}$) | $ln\ [(V_c/V)(T_c/T)^{3/2}]$ |
| 1 | Ar | 87.29 | 150.65 | 29.1 | 75.0 | 12.6 | 1.3656 | 1.7653 |
| 2 | $O_2$ | 90.18 | 154.78 | 28.1 | 73.37 | 13.2 | 1.3529 | 1.7700 |
| 3 | $PH_3$ | 185.7 | 325.0 | 45.8 | 125 | 21.3 | 1.4683 | 1.8436 |
| 4 | $H_2Se$ | 231.8 | 410.6 | 41.0 | 115 | 30.3 | 1.5542 | 1.8890 |
| 5 | CO | 81.64 | 132.85 | 35.4 | 92.17 | 9.5 | 1.2546 | 1.6873 |
| 6 | $OsO_4$ | 403.86 | 676 | 64.6 | 178 | 35.4 | 1.4112 | 1.7863 |
| 7 | $CS_2$ | 319.4 | 552 | 62.4 | 170 | 29.4 | 1.4481 | 1.8229 |
| 8 | $PBr_3$ | 446.5 | 711 | 108.5 | 294 | 25.9 | 1.3196 | 1.6947 |
| 9 | $SbCl_5$ | 445.1 | 690 | 146.5 | 389 | 19.4 | 1.2113 | 1.6341 |
| 10 | $GeCl_4$ | 356.25 | 552 | 124.1 | 330 | 17.3 | 1.2082 | 1.6349 |
| 11 | $CClF_3$ | 191.85 | 301.9 | 68.5 | 179.4 | 14.0 | 1.2217 | 1.6429 |
| 12 | $WF_6$ | 290.45 | 452.7 | 87.1 | 233.0 | 18.0 | 1.2177 | 1.6497 |
| 13 | $Ni(CO)_4$ | 315.45 | 473 | 135.9 | 355 | 13.3 | 1.1145 | 1.5678 |
| 14 | $n$-$C_5H_{12}$ | 309.2 | 469.6 | 118.2 | 304 | 14.2 | 1.1061 | 1.5715 |
| 15 | $C_6H_6$ | 353.25 | 562.1 | 96.0 | 259 | 20.9 | 1.2404 | 1.6892 |

TABLE II The values of $\sigma$ calculated by means of Eq. (22)

| | | $T(K)$ | | $V \times 10^6$ ($m^3 \times mol^{-1}$) | | $\sigma \times 10^3$ ($N \times m^{-1}$) | |
|---|---|---|---|---|---|---|---|
| *Ns.* | *Compound:* | *Boil.* | *Crit.* | *Boil.* | *Crit.* | *Calc.* | *Exper.* |
| 1 | $H_2$ | 20.397 | 33.19 | 28.47 | 65.0 | 2.4 | 2.5 |
| 2 | $F_2$ | 85.0 | 144.31 | 25.1 | 66.22 | 13.6 | 14.0 |
| 3 | $Br_2$ | 332.35 | 584 | 51.5 | 127 | 32.4 | 34.7 |
| 4 | $CH_4$ | 111.66 | 190.6 | 38.01 | 99.0 | 13.5 | 13.0 |
| 5 | $GeH_4$ | 184.7 | 308.0 | 55.6 | 150 | 17.3 | 17.7 |
| 6 | $B_2H_6$ | 180.7 | 290.0 | 63.1 | 173.1 | 15.0 | 14.7 |
| 7 | $H_2S$ | 212.87 | 373.3 | 35.9 | 99.7 | 29.4 | 29.6 |
| 8 | $SO_2$ | 263.1 | 430.65 | 43.9 | 122.0 | 28.9 | 28.5 |
| 9 | $ClO_3F$ | 226.5 | 368.4 | 59.9 | 161.0 | 19.4 | 20.0 |
| 10 | $PSCl_3$ | 398.0 | 622 | 116.4 | 313 | 20.6 | 21.1 |
| 11 | $BBr_3$ | 364.2 | 581 | 103.3 | 280 | 21.2 | 20.3 |
| 12 | $HgBr_2$ | 592.0 | 1011 | 74.2 | 209 | 49.3 | 50.3 |
| 13 | $CCl_4$ | 349.73 | 556.4 | 103.9 | 276 | 19.8 | 19.6 |
| 14 | $SiCl_4$ | 330.45 | 507.2 | 121.2 | 326.3 | 16.2 | 15.4 |
| 15 | $SnCl_4$ | 387.0 | 592.0 | 131.7 | 351.0 | 17.6 | 17.0 |
| 16 | $S_2Cl_2$ | 411.0 | 665 | 90.9 | 248 | 26.9 | 27.3 |
| 17 | $SF_4$ | 232.7 | 364.1 | 59.6 | 162 | 19.1 | 19.9 |
| 18 | $SF_6$ | 204.2 | 318.7 | 75.3 | 200.4 | 13.9 | 13.9 |
| 19 | $CF_4$ | 145.2 | 227.5 | 54.4 | 146.7 | 12.6 | 12.3 |
| 20 | $CCl_3F$ | 296.85 | 471.2 | 93.1 | 248.0 | 18.1 | 17.9 |
| 21 | $CBrCl_2F$ | 325.0 | 514 | 99.9 | 264 | 18.6 | 18.3 |
| 22 | $CHCl_2F$ | 282.07 | 451.6 | 75.0 | 197.0 | 19.8 | 20.1 |
| 23 | $C_2H_2$ | 189.2 | 308.3 | 42.3 | 113 | 20.4 | 19.6 |
| 24 | $C_2H_4$ | 169.35 | 282.4 | 49.2 | 129 | 16.7 | 16.5 |
| 25 | cyclo-$C_5H_{10}$ | 322.4 | 511.6 | 98.0 | 260 | 18.9 | 18.3 |
| 26 | $(C_2H_5)_2O$ | 308.75 | 466.7 | 106.5 | 280 | 15.6 | 15.7 |
| 27 | $C_6H_5Cl$ | 404.9 | 632.4 | 115.0 | 308 | 21.0 | 20.3 |

their normal boiling points $T$ were taken from [5, 7–17]. The comparison of the values of $\sigma$ calculated by means of Eq. (22) and the experimental ones shows that this equation describes the surface tension of all the investigated liquids adequately. The average absolute value (module) of its calculation error is equal to 2.6%. Equation (1) applied to the data set of Table II gives the average calculation error which is equal to 3.9%.

It is important to note that the above mentioned temperature limitation of Eq. (22) (which follows from the use of Eq. (3)) doesn't mean that the temperature of the investigated liquid may be only its boiling point or the nearest temperature. If the value of $T$ of this liquid is high enough in order to satisfy the temperature limitation caused by the application of Eq. (3), its surface tension will be described by means of Eq. (22). This conclusion may be illustrated on the basis of the data set of Table III. The liquids presented in Table III are

TABLE III The values of $\sigma$ calculated by means of Eqs. (22) and (1) for some chemical substances at the temperatures which are lower than their boiling points

| | | | | $V \times 10^6$ ($m^3 \times mol^{-1}$) | | $\sigma \times 10^3$ ($N \times m^{-1}$) | |
|---|---|---|---|---|---|---|---|
| *Ns.* | *Compound:* | *T(K)* | $T_c(K)$ | $V^a$ | $V_c$ | *Calc.* | *Exper.* |
| 1 | He | 1.55 | 5.189 | 29.37 | 57.31 | 0.383/0.816$^b$ | 0.354 |
| | | 3.05 | | 30.66 | | 0.286/0.463 | 0.224 |
| 2 | $N_2$ | 70.05 | 126.19 | 33.3 | 89.47 | 10.25/11.50 | 10.53 |
| 3 | $CH_4$ | 101.40 | 190.6 | 36.7 | 99.0 | 14.7/17.1 | 15.1 |
| 4 | $AsH_3$ | 180.15 | 375.0 | 45.8 | 128 | 26.1/32.3 | 26.6 |
| 5 | $CS_2$ | 293.15 | 552 | 60.3 | 170 | 31.8/35.7 | 32.4 |
| 6 | HBr | 192.75 | 363.0 | 36.4 | 100 | 28.6/32.9 | 27.8 |
| 7 | $SeF_4$ | 290.95 | 573 | 56.1 | 171.5 | 36.97/40.80 | 36.30 |
| 8 | $C_2H_6$ | 173.15 | 305.4 | 53.6 | 148 | 18.4/19.7 | 17.8 |
| 9 | $N(CH_3)_3$ | 194.15 | 433.2 | 78.4 | 254 | 23.2/27.7 | 25.2 |
| 10 | $C_6H_5Cl$ | 293.15 | 632.4 | 101.7 | 308 | 27.32/33.01 | 33.56 |
| 11 | $C_6H_5Br$ | 293.15 | 670.2 | 105.0 | 324 | 28.6/35.9 | 36.5 |
| 12 | $(C_6H_5)_2$ | 350.15 | 789 | 155.8 | 502 | 26.6/32.1 | 34.4 |

a – at the temperature *T*(K).

b – the values of $\sigma$ calculated by means of Eq. (1).

considered at the temperatures $T$ which are lower than their normal boiling points. Their experimental values $T_c$, $V_c$, $V$ at $T$ and $\sigma$ at $T$ were taken from [5, 7–17]. The comparison of the calculated values of $\sigma$ and the corresponding experimental ones (see Tab. III) allows to draw the following conclusion. In the case of the above mentioned lower temperatures, Eq. (22) is applicable for the adequate description of $\sigma$ of those low-boiling liquids, whose nonpolar (or low-polar) molecules are small and have the form which may be considered as nearly spherical. For these compounds Eq. (22) is more precisely than Eq. (1) (see Tab. III). However, the surface tension of the high-boiling compounds with their non-spherical molecules (see substances 10–12 in Tab. III) cannot be calculated by means of Eq. (22) at the considered temperatures which are far from their boiling points.

It is interesting that sometimes intermolecular interactions may decrease by passing to lower temperatures. In this case, the accuracy of Eq. (22) will be higher in this low-temperature region. So, for instance, superfluid helium II ($T = 1.55\,\mathrm{K}$) obeys Eq. (22) better than normal helium I ($T = 3.05\,\mathrm{K}$) does (see Tab. III).

## 4. CONCLUSION

It is to be noted that Eq. (22) cannot be used for such liquid chemical substances as, for instance, amines, alcohols, melts of metals and others. However, for non-associated near-boiling liquids (or for some liquids which are similar to compounds 1–9 in Tab. III) it allows one to calculate $\sigma$ more precisely than other equations proposed earlier.

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