MODIFICATION OF THE CELL MODEL AND THE EQUATION OF STATE OF LIQUIDS

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Using an effective intermolecular potential and a modification of the cell model, we obtain the equation of state for liquids. The parameters of the equation of state are calculated for 60 different materials.

The fundamental hypothesis of the cell model is that a liquid has a quasicrystalline structure and can be represented as a system of noninteracting cells, each containing a single particle. The self-consistent field principle is used, according to which the interaction energy of a given molecule is equal to the energies of the nearest neighbor cells to the given molecule. This model is usually applied to simple liquids composed of spherical molecules (atoms), and the Lennard–Jones (12-6) potential is normally used as the interaction potential.

Without dwelling here on the analytical apparatus of the theory, we note th at the theory (depending on a series of additional assumptions) leads to different equations of state of a liquid, such as the Lennard–Jones and Devonshire equation [1] and the Frenkel equation [2]. Unfortunately, these equations of state do not give a correct quantitative description of the thermal properties of even simple liquids [3]. For example, the disagreement between the calculated and experimental values of the density is of order $\delta\rho \approx$ 20% for argon. The error probably occurs because it is assumed in these theories [1, 2] that the structure of the liquid is invariable, and this contradicts the actual physical picture.

Unlike [1, 2], we assume that the structural characteristics of a liquid change with increasing temperature, as suggested by the experimentally observed temperatur dependence of the coordination number for simple materials [4]. This modification of the cell model leads to a more adequate representation of real liquids.

In addition, the pair potential is taken to be an effective (12-6) potential with parameters ε and σ that depend on temperature

$$U = 4\varepsilon(T)\left[\left(\frac{\sigma(T)}{r}\right)^{12} - \left(\frac{\sigma(T)}{r}\right)^{6}\right]. \tag{1}$$

The potential function (1) represents a generalized model in which the actual complicated interaction of the molecules is replaced by a centrally symmetric interaction which is approximated by a Lennard–Jones (12-6) potential with parameters ε and σ that depend on the temperature. This approach assumes that the potential (1) is universal and that it can be applied to a material with an arbitrary complicated interaction between molecules.

Without dwelling on the fundamental and applied aspects of the effective potential method (see [5, 6]), we note that the potential function (1) in combination with the (modified) cell model of a liquid can be used to obtain an equation of state for arbitrarily complicated materials, including polar liquids [7]:

$$Z = 1 - 1{,}744\,\frac{\varepsilon_\ell}{kT}\left[(b_{0\ell}\rho)^2 - 0{,}4654\,(b_{0\ell}\rho)^4\right]. \tag{2}$$

Each of the terms in (2) has a definite physical meaning. The first term corresponds to the ideal-gas state of the liquid (Z = 1), when the average distance between nearest neighbors is r_e. The second and third terms take into account the attractive and repulsive forces between molecules that arise when the distance between nearest neighbors is not equal to the equilibrium distance ($r \neq r_e$).

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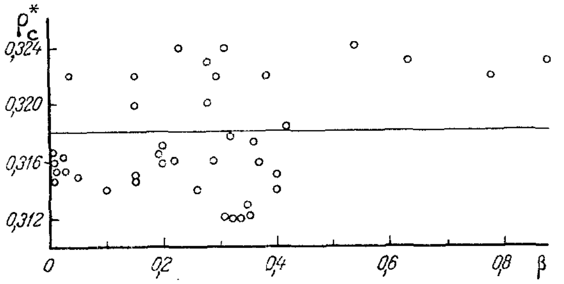

Fig. 1

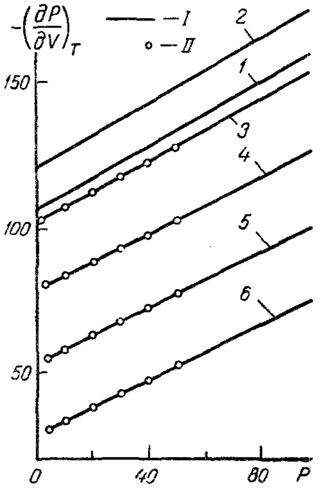

Fig. 2

Fig. 1. Critical density ρ_c^* for different liquids plotted against the Pittser acentricity factor β.

Fig. 2. Dependence of $(\partial p/\partial V)_T$ on pressure for water. I: calculated from the equation of state (2); II: experimental values of the derivative. 1) T = 273.15°K; 2) 323.15; 3) 373.15; 4) 423.15; 5) 473.15; 6) 523.15°K; P, MPa.

We note also that the density dependence on the right hand side of (4) is theoretically justified. The exponents of ρ (0, 2, 4) are determined by the lattice theory and the mathematical structure of the (12-6) potential, rather than empirically.

The parameters ε_ℓ and $b_{0\ell}$ in the equation of state reflect the structural characteristics of the liquid and the molecular interaction:

$$\varepsilon_\ell = \frac{\bar{Z}}{\bar{Z}_0}\varepsilon, \quad b_{0\ell} = \frac{\gamma_0}{\gamma} b_0; \; b_0 = 2\pi N\sigma^3/3. \tag{3}$$

It is evident from (3) that the temperature dependence of the parameters ε_ℓ and $b_{0\ell}$ comes from structural changes that occur in the liquid with increasing temperature (these structural changes are responsible for the temperature dependence of the coordination number $\bar{Z}$ and the structure factor γ) and also from the temperature dependence of the potential parameters ε(T) and σ(T). It is easy to see that the parameter ε_ℓ must decrease with increasing temperature T, since when the temperature increases from T_{tr} to T_c the intensity of the molecular interaction ε and the coordination number $\bar{Z}$ both decrease. It was shown in [8] that the temperature dependence of ε_ℓ found from experimental thermal data on liquids is essentially the same for all materials and ε_ℓ falls off exponentially with increasing T:

$$\frac{\varepsilon_\ell}{k} = \frac{\varepsilon_c}{k}\exp\left[c\left(1 - \frac{T}{T_c}\right)\right]. \tag{4}$$

We do not discuss the method of determining the parameters ε_ℓ and $b_{0\ell}$ from the thermal data on liquids, but note that the values of these parameters at $T = T_c$ can be used to determine the parameters of the effective potential (ε_c and σ_c) at the critical temperature of the material. This follows directly from (3).

It was shown in [8] that the following relation holds for all materials studied to within an error of less than a percent:

$$T_c = 1{,}26\,\frac{\varepsilon_c}{k}. \tag{5}$$

Hence the reduced critical temperature for real materials is $T_c^* = 1.26$, and this agrees with the result of [9] ($T_c^* = 1.29$).

The dimensionless critical density $\rho_c^* = \rho_c N\sigma_c^3$ was calculated for a large number of liquids. The relation $\rho_c^* = 0.318 \pm 0.006$ was satisfied for normal materials (nonpolar or weakly polar liquids).

TABLE 1. Parameters of the Equation of State and Quality of Fit to the Thermal Properties of Liquids

Material	T_c, K	$a \cdot 10^2$	$b \cdot 10^5$	c	P_{max}, MPa	$\Delta\tau$	$\delta\rho$, %
Ne	44,40	6,8285	51,874	2,09	100	0,56...1,00	0,19
Ar	150,65	3,9769	9,5857	2,17	100	0,56...1,00	0,06
Kr	209,39	3,2704	5,6910	2,17	100	0,55...1,00	0,10
Xe	289,75	2,5250	3,1292	2,18	100	0,56...1,00	0,13
N_2	126,20	3,3752	10,0980	2,39	100	0,52...1,00	0,15
O_2	154,58	4,1025	9,8950	2,19	100	0,36...1,00	0,08
CO_2	304,20	3,4923	4,7888	2,83	100	0,72...1,10	0,22
NH_3	405,55	4,7470	4,6447	1,89	100	0,49...1,10	0,10
CH_4	190,17	3,0127	5,7296	2,17	100	0,47...1,10	0,13
C_2H_6	305,33	2,1248	2,7044	2,52	100	0,31...0,98	0,10
C_3H_8	369,80	1,5844	1,7350	2,75	60	0,27...0,98	0,12
n- C_4H_{10}	425,16	1,2584	1,2088	3,04	50	0,35...0,96	0,14
n-C_6H_{14}	507,35	0,8963	0,7507	3,49	50	0,44...0,93	0,19
n- C_7H_{16}	540,15	0,7808	0,6178	3,64	50	0,51...0,95	0,17
n- C_8H_{18}	568,76	0,6935	0,5275	3,75	50	0,52...0,96	0,17
$C_{11}H_{24}$	640,08	0,5252	0,3813	4,00	50	0,47...0,90	0,28
$C_{13}H_{28}$	676,65	0,4499	0,3158	4,10	50	0,45...0,85	0,22
$C_{17}H_{36}$	734,15	0,3492	0,2334	4,24	50	0,44...0,79	0,18
$C_{20}H_{42}$	767,84	0,2975	0,1911	4,32	50	0,48...0,74	0,18
$C_{30}H_{62}$	846,54	0,2005	0,1208	4,46	50	0,44...0,68	0,21
$C_{40}H_{82}$	896,75	0,1517	0,8887	4,53	50	0,47...0,65	0,19
C_2H_4	282,35	2,3799	3,3055	2,43	100	0,39...1,00	0,11
C_2H_5Br	503,80	1,5613	1,2761	2,85	98	0,56...1,02	0,22
$C_4H_8O_2$ (methylpropionate)	530,60	1,2024	0,9804	3,31	50	0,57...1,13	0,20
$C_4H_8O_2$ (ethylacetate)	523,20	1,1609	0,8976	3,53	50	0,57...1,02	0,25
C_6H_6	562,60	1,2674	0,9459	2,89	60	0,56...1,20	0,15
C_6H_{12} (cyclohexane)	553,40	1,0527	0,8131	3,30	80	0,54...1,26	0,18
C_6H_{12} (1-hexene)	504,00	0,9440	0,8085	3,50	70	0,56...0,94	0,21
C_7H_6O	695,00	1,0449	0,6367	3,10	50	0,42...0,91	0,20
C_7H_8	591,75	1,0405	0,7358	3,14	50	0,29...1,09	0,19
C_7H_{14} (methylcyclohexane)	572,15	0,8797	0,6544	3,63	70	0,49...1,00	0,19
C_8H_{10} (n-xylene)	618,15	0,8903	0,6150	3,33	50	0,52...1,05	0,22
C_8H_{10} (o-xylene)	631,59	0,8998	0,5992	3,32	50	0,40...1,03	0,18
C_8H_{10} (m-xylene)	619,15	0,8903	0,6037	3,17	20	0,48...0,76	0,11
C_8H_{10} (ethylbenzol)	619,55	0,8978	0,6240	3,36	50	0,30...0,93	0,18
C_8H_{16} (1-octene)	566,60	0,7270	0,5797	3,83	71	0,50...0,88	0,21
C_9H_{12} (isopropylbenzol)	631,00	0,7814	0,5230	3,65	50	0,45...0,91	0,19
R 10	556,3	1,1684	0,8752	3,00	112	0,59...0,91	0,16
R 11	471,2	1,3079	1,1528	2,80	20	0,51...1,00	0,14
R 12	385,2	1,4971	1,6373	3,00	161	0,57...0,84	0,17
R 13	302,0	1,7712	2,4266	2,90	50	0,30...0,99	0,22
R 14	227,5	2,2160	3,9926	3,10	57	0,40...0,98	0,20
R 20	536,6	1,4245	1,1127	2,90	98	0,51...0,65	0,07
R 21	451,6	1,6560	1,5058	2,50	20	0,67...0,96	0,12
R 22	369,3	1,9935	2,2876	2,90	160	0,68...0,82	0,27
R 23	299,0	2,5108	3,4368	2,60	50	0,41...1,00	0,39
R 30	510,0	1,8229	1,4846	2,70	101	0,59...0,69	0,04
R 32	308,0	2,8239	3,2528	2,30	20	0,80...1,10	0,18
R 40	416,2	2,3574	2,2614	2,50	160	0,60...0,73	0,13
R 13Br1	340,2	1,6184	1,9488	3,01	50	0,50...0,90	0,09
R 152A	386,65	1,8899	2,0254	2,63	50	0,41...1,0	0,09
R 113	487,25	0,9970	0,8580	3,26	24	0,52...1,0	0,07
R 114	418,85	1,1138	1,1298	3,09	20	0,77...1,03	0,15
R 115	353,15	1,2450	1,4846	3,44	60	0,57...1,0	0,21
R 116	293,01	1,4399	2,0549	3,70	26	0,73...1,02	0,17
R 329	412,27	0,9332	1,0126	3,97	30	0,53...1,0	0,12
R 218	345,05	1,0734	0,1302	3,71	50	0,37...0,84	0,11
R C318	388,47	1,0437	1,2010	3,93	60	0,59...1,0	0,30
R 503	292,4	2,1067	2,9950	2,46	25	0,71...0,97	0,23

This is shown in Fig. 1, where the scatter of the values of ρ_c^* of real liquids from the average value $\rho_c^* = 0.318$ is plotted against the Pittser acentricity factor β. Each material is represented by a point in Fig. 1. The deviations are less than 2%, which is comparable to the error in determining the critical density ρ_c^*. Molecular dynamics calculations for the Lennard–Jones model give $T_c^* = 1.32\text{-}1.36$, $\rho_c^* = 0.32\text{-}0.36$ for the critical parameters [10].

An analysis of the equation of state (2) with the temperature-dependent parameters ε_ℓ and $b_{0\ell}$ shows that for temperatures $T_{tr} \le T \le T_c$ and densities $\rho > 1.8\rho_c$ it describes the thermal properties of liquids to within the experimental error of $\delta\rho = 0.1\text{-}0.3\%$.

It is of interest to see how well the equation of state (2) describes the thermodynamic behavior of liquids (particularly thermodynamic relationships not taken into account in

TABLE 2. Deviations between the Calculated Values on the Densities of Liquid Solutions and the Experimental Values [18]

$0,5C_{12}H_{26}+0,5C_{16}H_{34}$								$0,6C_{10}H_{22}+0,2C_{14}H_{30}+0,2C_{16}H_{34}$			
T=298,15 K		T=318,15 K		T=338,15 K		T=358,15 K		T=336,15 K		T=358,15 K	
P,MPa	$\delta\rho$, %	P,MPa	$\delta\rho$, %	P,MPa	$\delta\rho$, %	P,MPa	$\delta\rho$, %	P,MPa	$\delta\rho$, %	PMPa	$\delta\rho$, %
0,1	0,59	0,1	0,58	0,1	0,60	0,1	0,57	0,1	0,92	0,1	0,93
7,3	0,45	13,7	0,35	21,1	0,40	20,3	0,43	20,4	0,59	20,4	0,68
17,8	0,35	49,1	0,11	52,2	0,29	62,5	0,35	50,4	0,46	52,4	0,59
27,5	0,29	69,4	0,13	93,7	0,39	94,1	0,48	103,5	0,60	103,0	0,75
38,5	0,19	90,1	0,15	151,8	0,56	151,7	0,68	151,7	0,76	138,7	0,94
55,7	0,12	124,2	0,21	199,8	0,75	—	—	194,4	0,96	201,8	1,31

deriving the equation of state). An example is the linear variation of the derivative $(\partial P/\partial V)_T$ as a function of pressure (for T = const), which is true for all liquids, and has been experimentally verified.

The derivative $(\partial P/\partial V)_T$ as a function of pressure was calculated from the equation of state (2) for a large number of materials, including simple and polar liquids.

Figure 2, which is typical for the materials studied, illustrates the linear dependence of $(\partial P/\partial V)_T$ on pressure, and the good agreement with experiment. The points in Fig. 2 correspond to the experimental values of the derivative, obtained from data on water and the well-known thermodynamic relation

$$\left(\frac{\partial P}{\partial V}\right)_T = -\frac{W^2\rho^2}{\alpha},\ \alpha = \frac{C_p}{C_v}.$$

Hence we can write

$$-\left(\frac{\partial P}{\partial V}\right)_T = A(T) + B(T)P. \tag{6}$$

Equations were obtained, relating these coefficients with the parameters ε_ℓ and $b_{0\ell}$:

$$A = A(\varepsilon_\ell\ \ b_{0\ell}),\ B = B(\varepsilon_\ell,\ b_{0\ell}). \tag{7}$$

Explicit formulas are not presented here.

It is appropriate to point out that the linear dependence of the derivative $(\partial P/\partial V)_T$ on pressure is the basic assumption of the Tait equation of state, which is often used to calculate properties of liquids at high pressures [11]:

$$\left(\frac{\partial v}{\partial P}\right)_T = \frac{K}{P+L}. \tag{8}$$

Comparing (6), (7), and (8), we can see that the coefficients K and L in the Tait equation are functions of temperature and are uniquely determined by the structural characteristics of the liquid, as well as by the parameters of the intermolecular potential.

We consider the variation of density along the ideal gas curves. Using the equation of state (2) a relation can be established between the density ρ_{id} and the parameter $b_{0\ell}$ by setting the expression inside the square brackets in (2) equal to zero:

$$b_{0\ell}^{-1} = 0{,}6822\rho_{id}. \tag{9}$$

This relation shows that the parameter $b_{0\ell}$ can be given a macroscopic interpretation: it is proportional to the ideal-gas volume of the liquid. On the other hand, the parameter $b_{0\ell}^{-1}$ found from the experimental PvT-data varies linearly with temperature for all liquids studied. This supports the linear variation of the density along the ideal-gas curves, as found empirically for certain liquids in [12]. In addition, the linear dependence of the ideal-gas density on temperature also holds for strongly polar liquids, such as NH_3, freon R23 and R152A, benzaldehyde C_7H_6O with large dipole moments (μ = 1.47; 1.64; 2.23; 2.81D).

It then follows that the temperature dependence of the parameter $b_{0\ell}$ (or equivalently σ_ℓ^3 since $b_{0\ell} = 2\pi N\sigma_\ell^3/3$) has the form

$$\sigma_\ell^{-3} = a - bT. \tag{10}$$

Returning to (4), we obtain with the help of (5)

$$\varepsilon_\ell/k = 0{,}795 T_c \exp[c(1 - T/T_c)]. \qquad (11)$$

It follows from (10) and (11) that the equation of state (2) contains three independent constants (a, b, c) which can be determined from experimental PvT-data on liquids by performing a best fit to the thermal data [13]. These parameters were obtained using this method for a large number of materials and the equations of state were compared [13, 14]. The materials studied included the liquid phases of the inert gases, hydrocarbons, freon, inorganic compounds, and polar liquids.

Table 1 gives the parameters of the equation of state, the temperature and pressure regions for which the experimental data were obtained, and the errors in the density. These results show that the equation of state (2) is as accurate as the empirical equations of state. However, the advantage of (2) is that it has a theoretical basis, and its parameters have definite physical meanings. The extrapolation possibilities of the equation of state are good (reliable extrapolation into the high pressure and low temperature regions) and this supports the quasicrystalline approach, central to the model. Other features of the thermodynamic behavior of liquids were also established (for example, the linear variation of the enthalpy as a function of temperature along the ideal-gas curves).

Because (2) has a theoretical basis, one can attempt to use it for liquid mixtures to predict the properties of mixtures using only data on the pure components.

It is convenient to use the theory of conformal solutions, since the form of the effective potential is assumed to be the same for all liquid components (a Lennard–Jones (12-6) potential at a given value of the temperature). The one-liquid approximation turns out to be best in the framework of the theory of conformal solutions. In this approximation the solution is considered to be a homogeneous liquid in which the parameters ε and σ are given by the mixing rules [15]:

$$\varepsilon\sigma^3 = \sum_i \sum_j (\varepsilon_{ij}\sigma_{ij}^3)\, x_i x_j, \quad \sigma^3 = \sum_i \sum_j \sigma_{ij}^3 x_i x_j. \qquad (12)$$

In [7] mixing rules of the type (12) were used for the parameters ε_ℓ and σ_ℓ, as well as the mixing rules

$$\varepsilon\sigma^3 = \sum_i \sum_j (\varepsilon_{ij}\sigma_{ij}^3)\, x_i x_j, \quad \varepsilon = \sum\sum \varepsilon_{ij} x_i x_j, \qquad (13)$$

where the nondiagonal values of ε_{ij} and σ_{ij} are calculated according to the geometric mean and arithmetic mean rules of combination. A criterion for choosing the mixing rules (12) or (13) was given in [7]. We do not discuss this criterion here, but note that the correct description of the bulk properties of the mixtures is obtained with the help of this criterion. Analysis of a large number of binary, trinary, and multicomponent liquid mixtures shows that the equation of state (2) and the mixing rules (12) or (13) predict the properties of the solutions with a mean density error of $\delta\rho = 0.5\text{-}0.8\%$.

Table 2 shows that the densities of selected binary and trinary liquid mixtures, calculated using only data on the pure components agree closely with the experimental densities.

Because the equation of state (2) contains a total of three constants, it was used as the basis for the design of the thermophysical experiment of [16] and was used to predict the properties of liquids in the one-phase region [13]. As indicated above, the equation of state can also be used to predict the properties of solutions from data on the pure components [14, 17].

Finally, the mathematical simplicity of the equation of state, its compact form, and the presence of only three constants makes it suitable for use in automated information systems and in automated design systems.

NOTATION

$z = P/NkT\rho$; P, pressure; T, temperature; ρ, molar density; N, Avogadro's number; k, Boltzmann constant; ε and σ, potential parameters; z, coordination number; γ, structure factor; $\bar{z}_0$, γ_0, structural characteristics at the critical temperature; T_{tr}, triple-point temperature; T_c, critical temperature; ρ_c, critical density; W, thermodynamic speed of sound; x_i, molar fraction of the i-th component.

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DEPOSITION OF ULTRAFINE AEROSOLS IN A PLANE CHANNEL FROM A LAMINAR VAPOR–GAS FLOW OF NONUNIFORM CONCENTRATION

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The deposition of a high-dispersity aerosol in a water-vapor concentration gradient field in a thermal diffusion chamber with a control element [1] has been experimentally investigated.

One of the most complex problems of the separation of aerosol impurities from gas streams is the trapping of ultrafine aerosols with a particle size of less than $1 \cdot 10^{-6}$ m [2, 3]. In a number of studies [3-6] problems of this kind are solved by using the directional motion of the aerosol particles in the concentration gradient field of one of the components of the gas mixture on the basis of the fact that for fine aerosol particles the velocity of this motion, which is equal to the Stefan flow velocity and the rate of diffusiophoresis, depends only weakly on the size and shape of the particles, the physical properties of the material of which they are composed, and the phase transitions at their surface [7-9]. Since the rate of diffusiophoresis and the Stefan flow velocity are determined by the concentration gradient and the content of the active (diffusing) component of the gas mixture [7], in most cases various designs of thermal diffusion chamber (TDC) are used as the filter apparatus.

In order to improve the gas purification rate at high aerosol particle concentrations, the authors have proposed a filter model in the form of a TDC with a control element [1]. However, the study of the operation of this filter [10] is far from complete, since accurate

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