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Volume properties of solutions of oleic, linoleic, and linolenic acids in *n*-hexane and *n*-heptane at 298.15 K

A. G. Ramazanova,★ V. I. Yashkova, O. V. Balmasova, V. V. Korolev, and E. V. Ivanov

Institute of Solution Chemistry, Russian Academy of Sciences,
1 ul. Akademicheskaya, 153045 Ivanovo, Russian Federation.
Fax: +7 (493 2) 33 6237. E-mail: agr@isc-ras.ru

Densities of solutions of oleic, linoleic, and linolenic acids in *n*-hexane and *n*-heptane were measured using a vibrating-tube densimeter at 298.15 K in a concentration range of 0—0.012 molar fractions of solute. The measurement error does not exceed $\pm 5 \cdot 10^{-6}$ g cm^{-3}. The limiting partial molar volumes of fatty acids of the studied series in *n*-hexane and *n*-heptane and the excess volume properties of binary mixtures were calculated. On going from oleic to linolenic acid, the number of double bonds (>C=C<) in a solute molecule increases, the hydrocarbon chain length in a solvent molecule decreases, and compactness of the structure packing of the resulting solution increases. This is caused, as a whole, by the enhancement of the *n*-alkane—acid intermolecular interaction.

Key words: *n*-hexane, *n*-heptane; oleic, linoleic, and linolenic acids; density, volume characteristics of solutions.

Unsaturated fatty carboxylic acids (UFA) are active participants of metabolism and biochemical synthesis processes.[1—3] Oleic, linoleic, and linolenic fatty acids occupy an important place among the compounds of this class. The two last compounds are classified as essential acids of the vitamin F group, which are also used as adsorbates to provide aggregative and sedimentation stability of magnetic liquids.[4—8]

These UFA are interesting, first of all, due to the fact that their "long-chain" molecules contain the same number of carbon atoms ($C_{18:n}$) but differ in the number of double bonds $n_{C=C}$ in the *cis*-3-, *cis*-6-, and *cis*-9-positions with respect to the terminal methyl group.[1,2] Therefore, each of the compared UFA has specific features governing solvation in solutions.

The thermodynamic properties of UFA solutions in nonpolar organic solvents are of great interest, because these solutions form a basis for promising syntheses of highly dispersed magnetite-containing liquid-phase systems.[6—9]

Previously,[7] we studied the heats of solvation of oleic and linoleic acids in *n*-hexane to establish the solvating power of nonpolar solvents with respect to UFA. It is shown that the solvation process becomes less endothermic with an increase in $n_{C=C}$ in an acid molecule. These conclusions[7] can be supplemented by analysis of the data on the volume characteristics. In particular, the approach based on a comparison of the volume effects of solvation of fatty acids in aprotic nonpolar solvents of the same homological series gives useful information.

In the present work, we report the results of densimetric measurements of solutions of oleic, linoleic, and linolenic acids in two saturated hydrocarbons, namely, *n*-hexane and *n*-heptane, at $T = 298.15$ K.

Experimental

Experimental procedure. Hexane (pure) was purified according to a previously described procedure.[10] At the final stage, the purification product was distilled (excluding moisture), and the fraction boiling at 69 °C was collected. Heptane (reagent grade) was purified by fractionation distillation, collecting the condensate with the boiling point 98.5 °C. The overall content of admixtures in the purification products of *n*-alkanes (according to the analyses results) did not exceed 0.1 wt.%. To evaluate the purity of the solvents prepared for experiments, IR spectroscopy (on a Specord M-80 spectrophotometer in CaF_2 cells) and densimetry were used. The density (ρ_1) values of purified *n*-alkanes were 0.655195 (hexane) and 0.679620 g cm^{-3} (heptane), which agrees well with published results of precision (with the relative error at most 0.002% at 298.15±0.005 K) measurements of ρ_1: 0.65510,[11] 0.65525 [12] and 0.67965 [13], 0.67958 [14] g cm^{-3}, respectively.

Oleic (*cis*-9-octadecenoic) and linoleic (*cis*-6, *cis*-9-octadecadienoic) acids (pure) were purified according to recommendations[10] until the content of the main substance was not less than 99.9 wt.%. Linolenic (*cis*-3,*cis*-6,*cis*-9-octadecatrienoic) acid (Sigma Chem. Co.) with the content of the main substance above 99.5 wt.% was not additionally purified. The quality parameters of the UFA under study were evaluated by spectrophotometry: from the integral intensity of IR absorption bands in thin films on the KRS-5 glass plates. The densities of

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Table 1. Densities of solutions* ($\rho_{1,2}$/g cm^{-3}) of oleic, linoleic, and linolenic acids in hexane and heptane at 298.15 K

$c_{sm,2} \cdot 10^3$	$\rho_{1,2}$	$c_{sm,2} \cdot 10^3$	$\rho_{1,2}$	$c_{sm,2} \cdot 10^3$	$\rho_{1,2}$
Hexane—oleic acid		Hexane—linoleic acid		Hexane—linolenic acid	
4.0182	0.655241	3.0390	0.655223	3.4571	0.655239
11.794	0.655307	3.6618	0.655235	6.3511	0.655275
27.126	0.655515	24.243	0.655484	17.366	0.655412
36.561	0.655650	44.822	0.655751	18.813	0.655430
45.996	0.655743	69.582	0.656003	32.480	0.655600
58.967	0.655853	101.27	0.656437	48.558	0.655800
73.882	0.656041	117.29	0.656555	69.543	0.656061
79.020	0.656116	121.50	0.656651	121.48	0.656707
135.63	0.656738	205.21	0.657669	192.23	0.657587
202.44	0.657513	261.85	0.658316	244.84	0.658281
248.85	0.658060	318.44	0.659011	306.07	0.659003
302.61	0.658660	323.51	0.659053	361.46	0.659692
399.81	0.659841	380.94	0.659753	433.07	0.660542
492.23	0.660831	463.38	0.660685	553.36	0.662119
542.69	0.661409	545.80	0.661687	649.75	0.663278
Heptane—oleic acid		Heptane—linoleic acid		Heptane—linolenic acid	
3.1481	0.679647	3.4358	0.679652	3.6766	0.679641
4.5877	0.679659	5.7719	0.679672	4.9370	0.679660
8.2832	0.679702	7.0957	0.679695	6.1534	0.679676
22.256	0.679793	17.604	0.679765	9.8734	0.679713
38.146	0.679940	28.022	0.679856	15.267	0.679772
53.014	0.680067	33.807	0.679904	24.684	0.679859
82.832	0.680308	46.856	0.680063	31.596	0.679888
137.30	0.680779	79.539	0.680310	51.343	0.680109
212.06	0.681378	123.42	0.680690	78.988	0.680333
351.71	0.682543	154.57	0.680957	132.31	0.680803
433.49	0.683233	226.49	0.681601	226.11	0.681679
520.47	0.683972	356.92	0.682706	334.72	0.682709
545.98	0.684159	437.46	0.683453	453.09	0.683743
629.58	0.684927	532.86	0.684292	513.54	0.684306
707.95	0.685525	647.71	0.685243	667.85	0.685685

* The density values were obtained by averaging of results of several measurements (n = 4—5) at the given $c_{sm,2}$ values.

oleic, linoleic, and linolenic acids (ρ_2) at 298.15 K were 0.895593, 0.902707, and 0.917132 g cm^{-3}, respectively.

Solutions of the acids in n-hexane and n-heptane were prepared gravimetrically with the error not higher than $\pm 1 \cdot 10^{-5}$ g. The solute concentration was expressed in the solvomolality units*: $c_{sm,2} = 55.50843 \cdot x_2/(1 - x_2)$, where x_2 is the molar fraction of the acid. Since the n_2/n_1 ratio remains unchanged when the solvent is replaced, this concentration scale is convenient[15,16] for the estimation of the limiting partial (apparent) molar volumes $\bar{V}_2^\infty$ ($\equiv V_{\varphi,2}{}^\infty$) and for comparison of the molar volumes in different solvating media.

The density of solutions ($\rho_{1,2}$) was measured with the error not higher than $\pm 5 \cdot 10^{-6}$ g cm^{-3} (and reproducibility not higher than $\pm 2 \cdot 10^{-6}$ g cm^{-3} in a series of five measurements) using a specially designed precision vibrating-tube densimeter. The temperature of a measurement cell with a capacity of ~2 cm^3 was maintained constant with an accuracy (to $2 \cdot 10^{-3}$ K) to ensure reliability of the results. For structure details of the densimetric setup and the procedure of experimental measurements, see Refs 15 and 17. The results of measurements of $\rho_{1,2}$ are given in Table 1.

Experimental data processing. The regression analysis showed that the obtained dependences of $\rho_{1,2}$ on $c_{sm,2}$ are adequately described by the first-order equation $\rho_{1,2} = a_0 + a_1 c_{sm,2}$. The a_i coefficients (with standard deviations $\sigma_{0.95}$) are presented in Table 2.

The $\rho_{1,2}$ values approximated by this equation (at measured concentrations, see Table 1) were used for calculations of $\bar{V}_2^\infty$ ($\equiv V_{\varphi,2}{}^\infty$) and the excess molar volumes $V_{1,2}{}^E$ of solutions of UFA in n-alkanes. To estimate $\bar{V}_2^\infty$, the rational (or so-called "non-extrapolation") procedure[15,18] was used, which has an enhanced resistance of the apparent molar volume to the influence of errors on determining $\rho_{1,2}$ and $c_{sm,2}$ in the area of high dilu-

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* Solvomolality $C_{sm,2}$ is the dimensionless parameter of the composition of the solution expressed by the equation $c_{sm,2} = (n_2/n_1) \cdot 55.50843$, where n_1 and n_2 are the amounts of the solvent and solute, respectively.[15] The normalized factor 55.50843 is numerically equal to the amount of the substance (mol) H_2O in 1000 g of water.

Table 2. Coefficients a_0 and a_1 in the equation $\rho_{1,2} = a_0 + a_1c_{\mathrm{sm},2}$*

Solvent	Acid	a_0	a_1
		g cm^{-3}	
n-Hexane	Oleic	0.655198 ($7.6 \cdot 10^{-6}$)	0.011477 ($3.1 \cdot 10^{-5}$)
	Linoleic	0.655198 ($7.9 \cdot 10^{-6}$)	0.011905 ($3.1 \cdot 10^{-5}$)
	Linolenic	0.655196 ($6.2 \cdot 10^{-6}$)	0.012449 ($2.2 \cdot 10^{-5}$)
n-Heptane	Oleic	0.679619 ($5.7 \cdot 10^{-6}$)	0.008358 ($1.7 \cdot 10^{-5}$)
	Linoleic	0.679621 ($5.6 \cdot 10^{-6}$)	0.008713 ($2.1 \cdot 10^{-5}$)
	Linolenic	0.679621 ($5.3 \cdot 10^{-6}$)	0.009108 ($2.0 \cdot 10^{-5}$)

* The root-mean-square error of approximation is given in parentheses.

tions. According to this procedure, the dependence of the solution volume $V_{1,2} = (55.50843M_1 + c_{\mathrm{sm},2}M_2)/\rho_{1,2}$ on $c_{\mathrm{sm},2}$ (M_1 and M_2 are the molar weights of the components) was approximated by the following equation:

$$V_{1,2} - 55.50843V_1 = b_0(\equiv \bar{V}_2^{\infty} \equiv V_{\varphi,2}^{\infty})c_{\mathrm{sm},2} + b_1c_{\mathrm{sm},2}^2. \quad (1)$$

Here $V_1 = M_1/\rho_1$ is the molar volume of the solvent, and b_1 is the "slope parameter," $(V_{1,2} - 55.50843V_1)/c_{\mathrm{sm},2} = V_{\varphi,2}(c_{\mathrm{sm},2})$.

The main advantage of this method for processing of the dependences of $V_{\varphi,2}$ on $c_{\mathrm{sm},2}$ compared to the "traditional" extrapolation procedure is that the value of the section cut by the straight line on the ordinate ($V_{1,2}{}^{\infty} = 55.50843V_1$) is known with an accuracy higher than the accuracy of calculation of $V_{\varphi,2}$.[18]

The results of calculations by Eq. (1) and the volume characteristics of "pure" oleic, linoleic, and linolenic acids are presented in Table 3.

Results and Discussion

As can be seen from the data in Table 3, the structure packings of UFA in the individual state are fairly loose unconsolidated molecular aggregates ($V_2 \gg V_{\mathrm{w},2}$), with compactness increasing with an increase in $n_{\mathrm{C=C}}$. The difference between the limiting partial molar volumes of linolenic acid in n-hexane and n-heptane and the corresponding intrinsic molar volumes does not exceed 5.7%, whereas for a heptane solution of oleic acid the $\bar{V}_2^{\mathrm{E},\infty}$ value is close to zero (*i.e.*, $\bar{V}_2^{\infty} \approx V_2$). This indicates that the volumes of the free ("excluded") space near an acid molecule, which is surrounded by intrinsic molecules and nonpolar solvent molecules, are comparable.

At the same time, the regular increase in the negative $\bar{V}_2^{\mathrm{E},\infty}$ values in the series of oleic, linoleic, and linolenic acids indicates an increasing compression of the molecular packing of an acid—solvent solvate complex that formed. A similar tendency is observed when heptane is replaced by hexane in the solvate environment of a molecule of dissolved UFA (see Table 3).

The decrease in $\bar{V}_2^{\infty}$ with shortening of the hydrocarbon chain length in an n-alkane molecule is related, most likely, to the fact that on transition from heptane to hexane the structure packing of the solvent that surrounds the UFA molecule is loosened and its compressibility increases. This weakens the interaction between the solvent molecules.* It is most likely that the observed packing changes upon solvation of acids are directly related to differences in both the "solvate" properties of the solvating medium and the ability of "bulky" (long-chain) solute molecules to incorporate into the initial structural matrix of the solvent.

The UFA under study are dissolved in hexane with an endothermic effect, which becomes less pronounced with

Table 3. Volume properties (cm^3 mol^{-1}) of the studied unsaturated fatty acids in the individual state ($V_{\mathrm{w},2}$, V_2) and in their infinitely dilute solutions in n-alkanes ($\bar{V}_2^{\infty}$, $\bar{V}_2^{\mathrm{E},\infty}$) at 298.15 K

Solvent	Acid	$V_{\mathrm{w},2}{}^a$	$V_2{}^b$	$V_2/V_{\mathrm{w},2}$	$\bar{V}_2^{\infty}$	$-b_1$	$-\bar{V}_2^{E,\infty\ c}$
n-Hexane	Oleic	180.3	315.40	1.75	303.22	5.25	12.18
	Linoleic	177.3	310.68	1.75	295.37	5.30	15.31
	Linolenic	174.3	303.59	1.74	286.23	5.35	17.36
n-Heptane	Oleic	180.3	315.40	1.75	314.97	3.83	0.43
	Linoleic	177.3	310.68	1.75	307.73	3.90	2.95
	Linolenic	174.3	303.59	1.74	300.01	3.97	3.58

[a] The van der Waals volume $V_{\mathrm{w},2} = v_{\mathrm{w},2}N_{\mathrm{A}}$ was estimated by the proposed calculation method for $v_{\mathrm{w},2}$ of organic "overloaded" molecules (N_{A} is Avogadro´s number).[19]

[b] Molar volume of the acid: $V_2 = M_2/\rho_2$ (measured values of the ρ_2 density are given in Experimental).

[c] Limiting excess molar volume (or "volume effect of solvation")[20] of the acid: $V_2{}^{E,\infty} = \bar{V}_2^{\infty} - V_2$.

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* According to earlier published data,[21] the polarizability $\alpha_{0,1}$ and force constant ϵ_{11}/k of the Lennard-Jones pair potential (12—6) for hexane and heptane molecules are $11.83 \cdot 10^{-24}$ cm^3, 517 K and $13.70 \cdot 10^{-24}$ cm^3, 573 K, respectively. The evaporation enthalpies of the compared n-alkanes (in kJ mol^{-1}) are 31.55 (hexane) and 36.55 (heptane),[22] and the isothermic compressibilities $\beta_{\mathrm{T},1}$ are $16.27 \cdot 10^{-10}$ and $14.09 \cdot 10^{-10}$ Pa^{-1}, respectively.[23,24]

an increase in the $n_{C=C}$ number in a solute molecule. For instance, on going from oleic to linoleic acid, the enthalpy of solvation $\Delta_{sol}H_2^{csm,2}$ ($c_{sm,2} \approx 4.8 \cdot 10^{-4}$) in *n*-hexane decreases[7] from 8.8 to 6.2 kJ mol^{-1}. The negative $|\bar{V}_2^{E,\infty}|$ values increase simultaneously (see Table 3). However, when the $n_{C=C}$ number in an acid molecule increases and when heptane is replaced by hexane, the solvation enhances.

To verify this assumption, one can use model concepts making possible to divide the total (integral) thermodynamic characteristics into individual components. One of these approaches is the statistical mechanical calculation of the volume properties of the limiting dilute solution. The method is based on the approximation of molecules of components by hard spheres, in particular, in the framework of the scaled particle theory (SPT).

This approach[25—27] is based on the experimentally measured thermodynamic (including volume) properties of solvents, such as isothermic compressibility ($\beta_{T,1}$), whose expression is the exact solution of the Percus—Yevik state equation.[25] In addition, the procedure of imaginary replacement of the "real" polyatomic molecule by the solid sphere with the equivalent volume and diameter σ makes it possible to obtain data on relative volume changes upon the solvation of structural isomers or substances of the same homological series.

According to the SPT, the expression for the partial molar volume of the solute at infinite dilution has the form[27,28]

$$\bar{V}_2^{\infty} = \bar{V}_{cav}^{\infty} + \bar{V}_{int}^{\infty} + \beta_{T,1}RT, \qquad (2)$$

where $\bar{V}_{cav}^{\infty}$ and $\bar{V}_{int}^{\infty}$ are the contributions caused by the formation of the solvate cavity in the liquid medium of the solvent and intermolecular interactions 1—2, respectively; $\beta_{T,1}RT$ is the contribution taking into account the change in the standard state of the system during the gas → liquid transition.

The procedure of calculation of the volume contribution $\bar{V}_{cav}^{\infty}$ in Eq. (2) has been published earlier.[26—28] As can be seen from the data in Table 4, the positive volume contribution to $\bar{V}_2^{\infty}$ from the formation of the structural cavity in the solvent is compensated by the negative contribution $\bar{V}_{int}^{\infty}$, whose value (in the absolute expression) increases sharply on going from heptane to hexane and in the series of oleic, linoleic, and linolenic acids. The difference in volumes $|\bar{V}_{int}^{\infty}(C_6H_{14}) - \bar{V}_{int}^{\infty}(C_7H_{16})|$ on going from oleic to linolenic acid increases from 15.6 to 17.5 cm^3 mol^{-1}.

Taking into account the values of the "parameters of intermolecular interaction" of the compared solvents, the solvating power of the heptane medium should be higher. However, the incorporation of spatially extended UFA molecules into the more closely packed and less compressible structural matrix of heptane is fraught with additional difficulties of the configurational character, which are indicated, in particular, by an increase in $\bar{V}_2^{\infty}$ in the solvent (see Table 3). An increasing role of steric incompatibility for the formation of a UFA—*n*-alkane solvate complex is a probable reason for the weakening of intermolecular interactions 1—2 on going from hexane to heptane solutions of UFA.

Additional information on the packing effects in the solutions under study can be obtained from analysis of their excess molar volumes $V_{1,2}{}^E$. To estimate the latters, we used the formula[30,31] $V_{1,2}{}^E = x_2(V_{\varphi,2} - V_2)$, and the results of the corresponding calculations are shown in Fig. 1.

As can be seen from the data in Fig. 1, the increase in the UFA content in the solution results in the formation of more closely packed structures. In the series of oleic,

Table 4. Parameters of Eq. (2) and the volume characteristics (necessary for the calculation of the parameters) of the components of limiting dilute solutions of unsaturated fatty acids in *n*-alkanes at 298.15 K

Solvent	Acid	y^a	$\sigma_1 \cdot 10^8$ [b]	$\sigma_2 \cdot 10^8$ [c]	$\bar{V}_{cav}^{\infty}$	$-\bar{V}_{int}^{\infty}$
			cm		cm^3 mol^{-1}	
Hexane	Oleic	0.4295	5.637	8.300	319.6	20.4
	Linoleic	0.4295	5.637	8.254	315.2	23.9
	Linolenic	0.4295	5.637	8.207	310.8	28.6
Heptane	Oleic	0.4572	5.979	8.300	316.3	4.81
	Linoleic	0.4572	5.979	8.254	311.9	7.70
	Linolenic	0.4572	5.979	8.207	307.6	11.1

[a] Packing parameter estimated by the equation[27] $\beta_{T,1} = V_1(1-y)^4/[RT(1+2y)^2]$, where V_1 (cm^3 mol^{-1}) = 131.529 (hexane), 147.441 (heptane).

[b] Hard-sphere diameter of a solvent molecule calculated by the formula[26,27] $y = \pi\sigma_1 N_A/(6V_1)$, where N_A is Avogadro's number.

[c] Hard-sphere diameter of a solute molecule[29]: $\sigma_2 = [6V_{w,2}/(\pi N_A)]^{1/3}$.

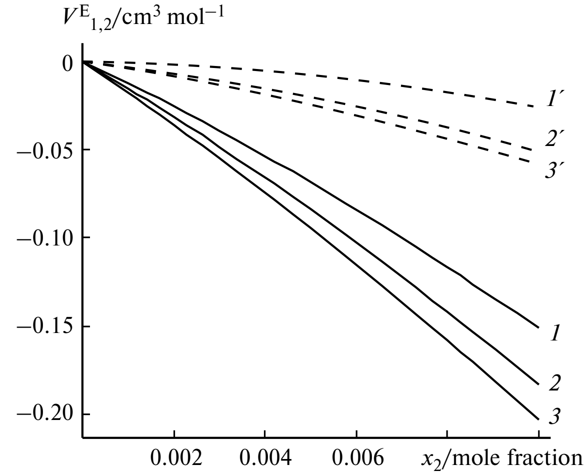

Fig. 1. Concentration plots of the excess molar volumes of *n*-alkane—unsaturated fatty acid mixtures for oleic (*1*, *1′*), linoleic (*2*, *2′*), and linolenic (*3*, *3′*) acids in hexane (*1—3*) and heptane (*1′—3′*) at 298.15 K.

linoleic, and linolenic acids and on going from heptane to hexane solutions of the acids, this tendency becomes more pronounced. When liquids with molecules differing both in chemical structure and geometric shape, are mixed, configuratives play the determining role in processes of structure packing formation and solvation of the dissolved component.[32,33]

Thus, when the number of double bonds (>C=C<) in acid molecules increases, their interaction with the solvent is enhanced and accompanied by the formation of more closely packed structures than those in the medium of a "pure" solute. On going from hexane to heptane, the solvating power (with respect to acids) of the solvent decreases, and this effect is caused by the crucial role of geometric (configurational) changes in the structure formation of an acid—solvent solvate complex. The nature of *n*-alkane exerts different effects on the character of the acid—solvent interaction. In particular, for the replacement heptane → hexane, the solvation of linolenic acid is enhanced to the greatest extent.

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