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ОБЪЕМНЫЕ СВОЙСТВА СМЕСИ АКРИЛОНИТРИЛ+ЭТАНОЛ В ИНТЕРВАЛЕ ТЕМПЕРАТУР (293,15-323,15) К ПРИ ВНЕШНЕМ ДАВЛЕНИИ

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Измерены плотности бинарной смеси акрилонитрила в этаноле во всем концентрационном диапазоне и в температурном интервале 293,15-323,15 К при внешнем давлении (87,9 кПа). Плотности растворов измерены с помощью колебательно-резонансного денсиметра Anton Paar DMA 4500 с точностью измерения плотности и температуры соответственно ±5·10^{-5} г/см^3 и ±0,03 К. Рассчитаны объемные параметры: избыточные молярные объемы, кажущиеся молярные объемы, парциальные и парциальные избыточные молярные объемы. Избыточные молярные объемы описаны уравнением Редлиха-Кистера. Параметры уравнения Редлиха-Кистера и стандартные отклонения избыточных молярных обьемов также рассчитаны. Величины избыточных молярных объемов отрицательны во всем концентрационном диапазоне в более низких температурах, но с ростом температуры от 293,15 К до 323,15 К отрицательные отклонения уменьшаются. Результаты интерпретируются на основе сильного взаимодействия между нитрильной группой акрилонитрила и ОН-группой этанола посредством водородной связи. Однако сигмоидальная форма зависимостей избыточных молярных объемов от мольных долей этанола от положительных до отрицательных значений указывает на различные взаимодействия в зависимости от состава бинарных смесей. Более того, такая форма зависимости избыточных молярных объемов от мольной доли при низких концентрациях этанола указывает на его самоассоциирующие действия, в результате которых в этих же концентрациях преобладают гомоассоциаты акрилонитрила с диполь-дипольным взаимодействием, а также гомоассоциаты этанола с водородными связями. В работе приведены новые данные по температурной зависимости плотностей и объемных свойств бинарного раствора акрилонитрил-этанол в температурном интервале 293,15-323,15 К. Полученные результаты сопоставлены с литературными данными.

Ключевые слова: акрилонитрил, этанол, денсиметрия, объёмные свойства

VOLUMETRIC PROPERTIES OF ACRYLONITRILE+ETHANOL MIXTURE OVER TEMPERATURE RANGE FROM (293.15 TO 323.15) K AT AMBIENT PRESSURE

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Densities of binary mixture of acrylonitrile in ethanol have been measured over the full range of compositions at temperatures from 293.15K to 323.15 K at the ambient pressure (87.9 kPa). The volumetric properties such as excess molar volumes, apparent molar volumes, partial molar volumes, excess partial molar volumes, standard partial molar volumes for binary mixture were calculated. Excess molar volumes were described by the Redlich–Kister polynomial equation. The Redlich–Kister coefficients and the standard deviations were calculated as well. The excess molar volumes exhibit mainly negative deviations from the ideal behavior over the whole range of composition for acrylonitrile+ethanol binary mixtures at low temperatures and become less negative

with increasing temperature from 293.15 to 323.15 K. The results are interpreted on the basis of strong interaction between group of acrylonitrile and OH group of ethanol by hydrogen bonding. However, the sigmoid shape of plots of excess molar volumes versus molar fraction of ethanol from positive to negative values indicates to the varying interactions depending upon the composition of binary mixtures. Moreover, it is also attributed to the self-associating effect of ethanol therefore the self-associates of acrylonitrile by dipole-dipole interactions and the self-associates of ethanol by hydrogen bonding between same molecules at low concentrations predominate too.

Key words: acrylonitrile, ethanol, densitometry, volumetric properties

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INTRODUCTION

Acrylonitrile (AN) as a polar solvent serves as a monomer to synthesize polyacrylonitrile (PAN). A fundamental understanding of the mixing behavior of AN with different solvents is therefore important from a technical and engineering viewpoint. On the other hand, the study of solutions of nitrile-containing compounds, such as AN, acetonitrile, etc is of great interest due to form both homo- and heteromolecular associates conditioned with their structure. Obviously, electronic effects (inductive and mesomeric) of alkyl groups in nitriles are responsible for the polarity of nitrile group and hence significant intermolecular interaction between –C≡N and other polar groups occur. To explain these phenomena, it should be mentioned that when groups such as C≡N are adjacent to a multiple bond, they withdraw π electrons from multiple bond through resonance or mesomeric effect. Obviously, such a type interaction occurs between *C=C* and C≡N groups in AN molecule (CH_2=CH-C≡N). As a result the polarity of C≡N group in AN molecule is increasing and hence more strong interaction between C≡N groups of AN and other polar groups takes place too.

It should be noted that the interaction of some solvent molecules with both PAN and AN takes place due to dipole-dipole interaction between nitrile groups and polar groups of solvents [1-9]. Moreover, according to [10] the dipole-dipole interaction between –C≡N group with other polar groups mainly leads to the formation of the intermolecular associate as the antiparallel dipolar conformation. It should be also noted that such interaction between C≡N group and other polar groups leads to the negative values of excess molar volumes of solutions as well [5-13]. On the other hand, there are several studies [14-20] for capability of a nitrile group formed heteromolecular associates by hydrogen bonding.

In this work, a detailed study of volumetric properties for AN in ethanol (EtOH) has been done through densitometry to elucidate the effect of the medium and particularly the role of temperature on the specific interactions between unlike molecules. This work also reports new data of densities and volumetric properties of AN+EtOH mixture over the full range of compositions at temperatures from 293.15K to 323.15 K. The excess molar volumes, V_m^E, apparent molar volumes, $V_{\phi,i}$, partial molar volumes, $\overline{V}_i$, excess partial molar volumes, $\overline{V}_i^E$ standard partial molar volumes, V_i^0, and thermal expansion coefficients, α_p, have been calculated as a function of composition. A Redlich-Kister type equation correlates the excess molar volumes.

EXPERIMENTAL METHOD

AN (vinyl cyanide; IUPAC name Prop-2-enenitrile) was purchased from Aldrich Chemical Co. with > 99% purity and content of 35-45ppm monomethyl ether hydroquinone as inhibitor (CAS No 107-13-1; C_3H_3N; MW 53.06 g/mol; mp:-83 °C; bp: 77 °C).

EtOH was purchased from Aldrich Chemical Co (CAS No 64-17-5; C_2H_6O; MW 46.07 g/mol; mp: -114 °C; bp: 78 °C; assay: > 99.8%) and was used without further drying or purification.

The densities of solutions were measured on an Anton Paar DMA 4500 vibrating-tube densitometer in the temperature range of (293.15-323.15) K and at ambient pressure (87.9 kPa). The accuracy of the density and temperature measurements was $\pm 5 \cdot 10^{-5}$ g/cm^3 and ±0.03 K, respectively. The densitometer was calibrated with dry air and double-distilled water. All solutions were prepared gravimetrically using a Sartorius CPA6235 balance with uncertainty of $\pm 1 \cdot 10^{-3}$ g.

RESULTS AND DISCUSSION

From the experimental values of the densities volumetric properties: excess molar volume, V_m^E, apparent molar volume, $V_{\phi,1}$, partial molar volume, $\overline{V}_i$, excess partial molar volume, $\overline{V}_i^E$ standard partial molar volume, V_i^0, and thermal expansion coefficient, α_p, were estimated.

Densities and Excess Molar Volumes

The experimental values of densities of the binary solution of AN in EtOH at the temperature range from (293.15 to 323.15) K and at ambient pressure (87.9 kPa) are presented in Table 1.

Table 1

Densities of AN(1)+EtOH(2) binary solution at T = (293.15-323.15) K

***Таблица 1*. Плотности бинарного раствора AN(1)+EtOH(2) при температурах T = (293,15-323,15) K**

X_2	ρ, g·cm^{-3}						
	293.15K	298.15K	303.15K	308.15K	313.15K	318.15K	323.15K
0.000	0.80643	0.80079	0.79516	0.78949	0.78379	0.77806	0.77228
0.103	0.80471	0.79923	0.79366	0.78805	0.78240	0.77671	0.77097
0.202	0.80347	0.79807	0.79257	0.78703	0.78145	0.77583	0.77015
0.303	0.80221	0.79690	0.79150	0.78606	0.78057	0.77503	0.76944
0.399	0.80095	0.79569	0.79040	0.78505	0.77967	0.77423	0.76874
0.502	0.79961	0.79453	0.78936	0.78415	0.77889	0.77357	0.76820
0.601	0.79833	0.79339	0.78836	0.78328	0.77815	0.77297	0.76773
0.701	0.79682	0.79203	0.78715	0.78223	0.77725	0.77222	0.76714
0.800	0.79516	0.79052	0.78579	0.78101	0.77619	0.77132	0.76639
0.900	0.79301	0.78856	0.78402	0.77944	0.77481	0.77014	0.76541
1.000	0.78968	0.78543	0.78111	0.77676	0.77238	0.76794	0.76346

The expanded uncertainties do not exceed U(ρ) = 1·10^{-4} g·cm^{-3} ,U(T) = 0.03 K, and U(p) = 0.5 kPa (level of confidence = 0.95)
Расширенные неопределенности не превышают U(ρ) = 1·10^{-4} г·см$^{-3}$,U(T) = 0.03 K, and U(p) = 0.5 кПа (доверительная вероятность = 0.95)

Excess molar volumes, V^E, of the above mentioned mixtures were calculated from the experimentally measured densities at each temperature using the following equation

$$V_m^E = (x_1M_1 + x_2M_2)/\rho - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (1)$$

where M_1 and M_2 represent the molar masses of AN and EtOH respectively. The x_1 and x_2 are molar fractions of AN and EtOH having densities ρ_1 and ρ_2, respectively.

As it follows from these data (Fig. 1) the excess molar volumes are mainly negative over the whole range of composition for AN + EtOH mixtures at low temperatures. These results are in agreement with those obtained earlier for AN in EtOH at 298.15 K [14]. To explain this observation it should be mentioned that the polarity of C≡N group in AN molecule is increasing due to mesomeric effect and hence more strong interaction between C≡N group of AN and *OH* group of EtOH by hydrogen bonding takes place. However the values of excess molar volumes become less negative with increasing temperature from 293.15 to 323.15 K. Moreover, those become even positive in concentration range richer by AN implying that the intermolecular forces between different molecules in those systems with increasing temperature become less strong. On other hand the S-shaped plots of V^E versus molar fraction of EtOH from positive to negative values are observed indicating the varying interactions depending upon the composition of binary mixtures. Moreover, it is also attributed to the self-associating effect of EtOH at low concentrations, therefore the self-associates of AN by dipole-dipole interactions between molecules predominate too.

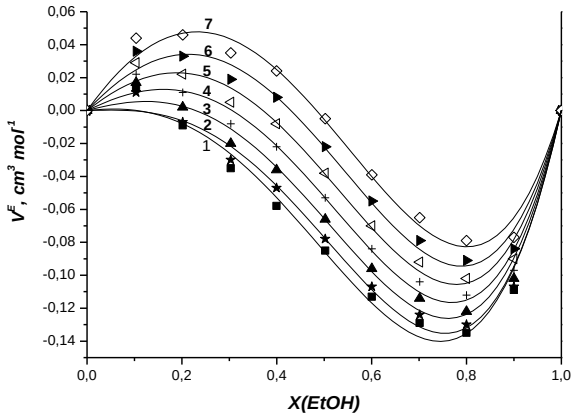

Fig. 1. Excess molar volumes of AN+EtOH binary solutions at 293.15K (1), 298.15K (2), 303.15K (3), 308.15K (4), 313.15K (5), 318.15K (6), 323.15K (7)
Рис. 1. Избыточные молярные обьемы бинарного раствора AN+EtOH при температурах 293,15 K (1), 298,15 K (2), 303,15 K (3), 308,15 K (4), 313,15 K (5), 318,15 K (6), 323,15 K (7)

Excess molar volumes were described by the Redlich–Kister polynomial equation [11, 21]:

$$V^E = x_1(1-x_1)\sum_{i=0}^{m} A_i(2x_1-1)^i \quad (2)$$

The Redlich–Kister coefficients, A_i, and standard deviations, σ, are summarized in Table 2. The standard deviations, σ, are evaluated from the equation:

$$\sigma = \left[\sum\left(V^E_{\text{expt}} - V^E_{\text{calc}}\right)^2/(m-n)\right]^{1/2} \quad (3)$$

where m is the number of results and n is the number of parameters retained in equation (3).

Table 2

Redlich-Kister fitting coefficients, Ai, and standard deviations, σ(V_m^E/ cm^3 mol^{-3}), of the excess molar volumes for the AN solutions in EtOH at temperatures from (293.15 to 323.15) K

***Таблица 2*. Параметры уравнения Редлиха-Кистера и стандартные отклонения избыточных молярных обьемов раствора AN в EtOH в температурном интервале T = (293,15-323,15) K**

T/K	A_0	A_1	A_2	$\sigma(V_m^E)$
293.15	-0.3419	-0.6645	-0.2929	0.008
298.15	-0.3081	-0.6555	-0.3419	0.007
303.15	-0.2625	-0.6587	-0.3217	0.007
308.15	-0.2096	-0.6617	-0.3132	0.007
313.15	-0.1521	-0.6650	-0.2892	0.007
318.15	-0.0884	-0.6687	-0.2778	0.007
323.15	-0.0217	-0.6752	-0.2504	0.055

Apparent and Partial Molar Volumes

In a binary liquid mixture, the apparent molar volumes $V_{\phi,1}$ and $V_{\phi,2}$ of components 1 (AN) and 2 (EtOH) are calculated by following equations [5, 11, 21]:

$$V_{\phi,1} = V_1^* + V_m^E / x_1 \quad (4)$$

$$V_{\phi,2} = V_2^* + V_m^E / x_2 \quad (5)$$

where V_1* and V_2* are the molar volumes of pure AN and EtOH, respectively (Table 3) which calculate from corresponding values of density. It should be noted that the apparent molar volumes are not constants (even at a given temperature), but are functions of the composition. The data of apparent molar volumes for the above mentioned binary solution at temperature from 293.15 K to 323.15 K are presented in Table 4.

Table 3

Molar volumes, V_m, of AN and EtOH at different temperatures

***Таблица 3.* Молярные обьемы AN и EtOH при разных температурах**

T,K	V_m	
	AN	EtOH
293.15	65.80	58.34
298.15	66.26	58.66
303.15	66.73	58.98
308.15	67.21	59.31
313.15	67.70	59.65
318.15	68.20	59.99
323.15	68.71	60.34

Table 4

Apparent molar volumes, $V_{\phi,i}$, and partial molar volumes, $\overline{V_i}$, of AN(1)+EtOH(2) binary solution at T=(293.15-323.15) K

***Таблица 4.* Кажущиеся и парциальные молярные обьемы бинарной смеси AN(1)+EtOH(2) в температурном интервале T = (293,15-323,15) K**

X_2	$V_{\phi,1}$	$V_{\phi,2}$	$\overline{V_1}$	$\overline{V_2}$
1	2	3	4	5
T = 293.15 K				
0.103	65.81	58.47	65.89	58.44
0.202	65.79	58.30	65.86	58.22
0.303	65.75	58.22	65.77	58.12
0.399	65.70	58.20	65.67	58.09
0.502	65.63	58.17	65.54	58.09
0.601	65.51	58.15	65.41	58.12
0.701	65.36	58.16	65.26	58.18
0.800	65.12	58.17	65.05	58.24
0.900	64.70	58.22	64.67	58.29
T = 298.15K				
0.103	66.27	58.77	66.35	58.75
0.202	66.25	58.62	66.31	58.56
0.303	66.22	58.56	66.23	58.46
0.399	66.18	58.54	66.14	58.43
0.502	66.10	58.50	66.02	58.42
0.601	65.99	58.48	65.89	58.44
0.701	65.85	58.48	65.76	58.49
0.800	65.61	58.49	65.55	58.56
0.900	65.19	58.54	65.17	58.61
T = 303.15 K				
0.103	66.75	59.14	66.82	59.11
0.202	66.73	58.99	66.79	58.92
0.303	66.70	58.92	66.70	58.81
0.399	66.67	58.89	66.62	58.77
0.502	66.60	58.85	66.50	58.75
0.601	66.49	58.82	66.37	58.77
0.701	66.35	58.82	66.24	58.82
0.800	66.12	58.83	66.05	58.88
0.900	65.71	58.87	65.68	58.94
T = 308.15 K				
0.103	67.23	59.53	67.30	59.49
0.202	67.22	59.37	67.27	59.28
0.303	67.20	59.28	67.19	59.16
0.399	67.17	59.26	67.11	59.12
0.502	67.10	59.21	66.99	59.09
0.601	67.00	59.17	66.87	59.11
0.701	66.86	59.16	66.74	59.16
0.800	66.65	59.17	66.56	59.22
0.900	66.24	59.20	66.20	59.27
T=313.15 K				
0.103	67.73	59.93	67.79	59.87
0.202	67.72	59.76	67.76	59.65
0.303	67.70	59.66	67.69	59.52
0.399	67.68	59.63	67.61	59.48
0.502	67.62	59.57	67.49	59.44

1	2	3	4	5
0.601	67.52	59.53	67.37	59.45
0.701	67.39	59.52	67.25	59.50
0.800	67.19	59.52	67.08	59.56
0.900	66.80	59.55	66.75	59.60
T = 318.15 K				
0.103	68.24	60.34	68.29	60.28
0.202	68.24	60.16	68.27	60.04
0.303	68.22	60.06	68.19	59.90
0.399	68.21	60.01	68.11	59.84
0.502	68.15	59.95	68.00	59.80
0.601	68.06	59.90	67.89	59.81
0.701	67.93	59.88	67.77	59.85
0.800	67.74	59.88	67.62	59.91
0.900	67.35	59.90	67.29	59.95
T=323.15 K				
0.103	68.75	60.77	68.80	60.69
0.202	68.76	60.57	68.78	60.43
0.303	68.76	60.46	68.71	60.27
0.399	68.75	60.40	68.64	60.21
0.502	68.69	60.33	68.53	60.17
0.601	68.61	60.28	68.42	60.17
0.701	68.49	60.25	68.30	60.21
0.800	68.31	60.24	68.17	60.27
0.900	67.94	60.26	67.86	60.30

The uncertainties are U(ρ) = $1\cdot10^{-4}$ g·cm^{-3}, U(V) = 0.5 cm^3·mol^{-1} or less, U(T) = 0.03 K, U(p) = 0.5 kPa (level of confidence = 0.95)
Погрешности U(ρ) = $1\cdot10^{-4}$ г·см$^{-3}$, U(V) = 0.5 см^3·моль$^{-1}$ or less, U(T) = 0.03 K, U(p) = 0.5 кПа (доверительная вероятность = 0.95)

The partial molar volumes, $\overline{V}_i$, can be determined from excess molar volumes using following equation:

$$\overline{V}_i = V_i^* + V^E / x_i + x_i(1-x_i)(\partial(V^E / x_i)/\partial x_i)_{T,P} \qquad (6)$$

where $(\partial(V_m{}^E/x)/\partial x_i)_{T,P}$ is calculated using Redlich–Kister polynomial equation (2) and V_i^* represent the molar volumes of the component 1 (AN) or 2 (EtOH). As it follows from presenting data, with the increasing of one's own mole fraction and temperature, the apparent molar volumes of AN in EtOH increase. The same behavior is mainly observed for partial molar volumes as well (Table 4).

At infinite dilution, an apparent molar volume and the corresponding partial molar volume coincide numerically becoming to be the standard quantity [22, 23]. Hence, the standard partial molar volumes, V_i^0, are considered of particular interest because of their usefulness in examining solute-solvent interactions, as solute-solute interactions can be assumed to be eliminated at infinite dilution.

The standard partial molar volumes, V_i^0, were estimated by extrapolating the apparent molar volumes to infinite dilution. The standard partial molar volumes of both components in binary mixture at temperatures from 293.15 K to 323.15 K are given in Fig. 2.

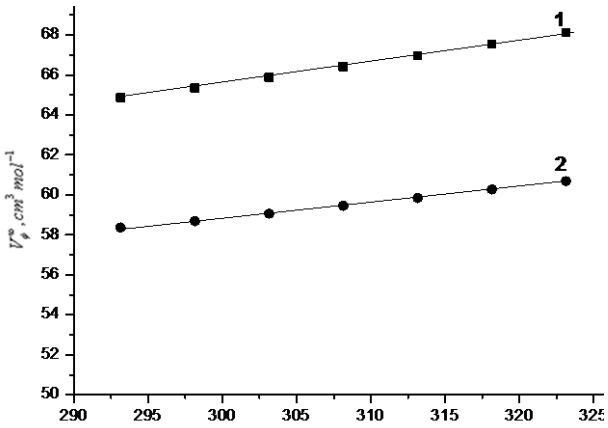

Fig. 2. The standard partial molar volumes, $V_i°$, of components of AN+EtOH binary mixture at temperatures from 293.15 to 323.15 K: AN (1); EtOH (2)
Рис. 2. Стандартные парциальные молярные объёмы компонентов смеси AN+EtOH в температурном интервале 293,15K-323,15 K: AN (1); EtOH (2)

As seen from data the infinite dilution values increase with increasing temperature. Noteworthy is the item that at infinite dilution the partial molar volumes and apparent molar volumes of both AN and EtOH are mainly smaller than for the corresponding pure solvents (Table 3 and 4). This also indicates the existence of interaction between these molecules.

The partial excess molar volume, $\overline{V}_i^E$, of a component in a AN+EtOH mixture has a physically justified, meaning of the "volume effect of dissolution" [24]. The values of $\overline{V}_i^E$ can be determined from the relation (7) and given in Fig. 3:

$$\overline{V}_i^E = \overline{V}_i - V_i^* \qquad (7)$$

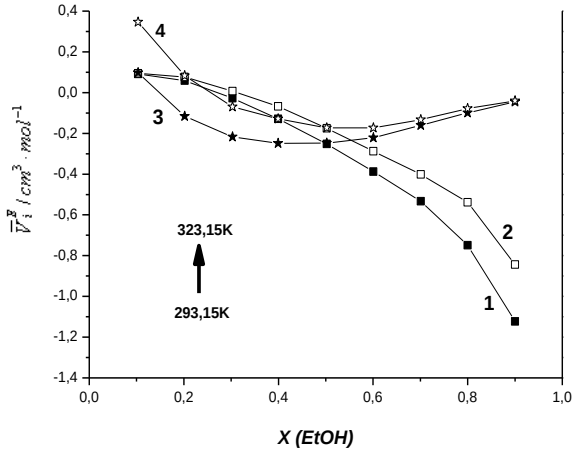

Fig. 3. Partial excess molar volumes, $\overline{V}_i^E$, of components of AN-EtOH binary solution at temperatures from (293.15 to 323.15) K: AN – 293.15 K (1) and 323.15 K (2); EtOH – 293.15 K (3) and 323.15 K (4)
Рис. 3. Парциальные избыточные молярные обьемы компонентов бинарной смеси AN-EtOH при температурах (293,15 – 323,15) K: AN – 293,15 K (1) и 323,15 K (2); EtOH – 293,15 K (3) и 323,15 K (4)

As seen from data, the values of partial excess molar volumes of individual components of this mixture increase with increasing temperature. Noteworthy is the item that in mixture apparent and partial volumetric properties of EtOH have weakly expressed exponential dependence from own molar fraction.

CONCLUSIONS

The excess molar volumes exhibit mainly negative deviations from the ideal behavior over the whole range of composition for AN+EtOH mixtures at low temperatures and become less negative with increasing temperature from 293.15 to 323.15K. The results are interpreted on the basis of strong interaction between C≡N group of AN and OH group of EtOH by hydrogen bonding. However the sigmoid shape of plots excess molar volumes of versus molar fraction of EtOH from positive to negative values indicates to the varying interactions depending upon the composition of binary mixtures. Moreover, it is also attributed to the self-associating effect of EtOH, therefore the self-associates of AN by dipole-dipole interactions between molecules at low concentrations predominate too.

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