Density and Viscosity of Acrylonitrile + Cinnamaldehyde, + Anisaldehyde, and + Benzaldehyde at (298.15, 308.15, and 318.15) K

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The excess molar volume V^E and viscosity deviations $\Delta\eta$ have been derived from density ρ and viscosity η measurements of the binary mixtures of acrylonitrile with cinnamaldehyde, anisaldehyde, and benzaldehyde measured at (298.15, 308.15, and 318.15) K. The data have been correlated with the Grunberg−Nissan equation. Moreover, deviations in isentropic compressibility ΔK_S have been calculated from ultrasonic speed measurements of these binary mixtures at 298.15 K. These results were fitted to the Redlich−Kister polynomial equation.

Introduction

Studies on thermodynamic and transport properties are important in understanding the nature of molecular interactions in binary liquid mixtures. Properties of mixtures are useful for many types of transport and process equipment in the chemical industry.[1–5] The study of mixtures of acrylonitrile in various solvents is of interest because of its wide use as an important industrial monomer for polyacrylonitrile as well as for investigating the effect of the simultaneous presence of the C═C double bond and the polar nitrile C≡N group on the molecular interactions. Further, the properties of liquid mixtures are also very important for polymer solubility, as some of the polymers which are insoluble in a single solvent can be dissolved in a solvent mixture. This behavior depends not only on the interactions between the polymer and each one of the liquids but also on the interactions between the liquids themselves. In fact, these interactions are decisive in determining the solubilization of the polymer and the coiling expansion of the polymer chains in solution.[6]

In the present paper, we report densities, viscosities, and ultrasonic speeds for the binary systems of acrylonitrile + cinnamaldehyde, acrylonitrile + anisaldehyde, and acrylonitrile + benzaldehyde at the temperatures of (298.15, 308.15, and 318.5) K and at atmospheric pressure over the entire composition range. The experimental data are used to calculate excess molar volumes, V^E, viscosity deviations, $\Delta\eta$, and deviations in isentropic compressibility, ΔK_S, of the mixtures. These results are useful for the interpretation of the nature of interactions that occur between acrylonitrile and the aromatic aldehydes. In addition, this work also provides a test of various empirical equations to correlate viscosity and ultrasonic sound data of binary mixtures containing polar components.

Experimental Section

Materials. High purity spectroscopic and analytical grade samples of 2-propenenitrile (acrylonitrile), 3-phenyl-2-propenal (cinnamaldehyde), 4-methoxybenzaldehyde (*p*-anisaldehyde), and benzaldehyde were purchased from S.D. Fine Chemicals Ltd., Mumbai, India. Acrylonitrile was washed with dilute H_2SO_4, then with dilute Na_2CO_3 and water, and again dried over anhydrous $CaCl_2$ and distilled fractionally.[7] Benzaldehyde was washed with 10 % Na_2CO_3 (until no more CO_2 evolved), then with saturated Na_2CO_3 and water, followed by drying with $CaCl_2$.[7] Anisaldehyde was washed with $NaHCO_3$, then with water, dried with anhydrous $MgSO_4$, and distilled under reduced pressure under N_2.[7] Cinnamaldehyde was crystallized from benzene and dried at 60 °C under vacuum.[7] The gas−liquid chromatography (GLC) analyses of these liquids indicated a mole fraction purity of > 99.0 %. The purity of the liquids was also checked by measuring their densities and viscosities at 298.15 K and was in reasonable agreement with the literature values as shown in Table 1.

Table 1. Comparison of Density *ρ*, Viscosity *η*, and Sound Speeds *u* with Literature Data at the Experimental Temperatures

		$\rho\cdot10^{-3}/(kg\cdot m^{-3})$		η/(mPas)		$u/(m\cdot s^{-1})$	
pure solvent	*T*/K	exptl	lit.	exptl	lit.	exptl	lit.
acrylonitrile	298.15	0.7943	0.8002[22]	0.3439	0.344[22]	1172.5	
	308.15	0.7879	0.7893[22]	0.3075	0.309[22]		
	318.15	0.7828		0.2838			
cinnamaldehyde	298.15	1.0459		3.4778		1540.2	
	308.15	1.039		2.6878			
	318.15	1.0323		2.2958			
anisaldehyde	298.15	1.1202		3.7055		1523.7	
	308.15	1.1117		2.8295			
	318.15	1.1049		2.2321			
benzaldehyde	298.15	1.0409		1.3923		1460.5	
	308.15	1.0335		1.2241			
	318.15	1.0266		1.1202			

Apparatus and Procedures. The densities were measured with an Ostwald−Sprengel-type pycnometer having a bulb volume of 25 cm^3 and an internal diameter of the capillary of about 0.1 cm, calibrated at (298.15, 308.15, and 318.15) K with doubly distilled water. The pycnometer with the test solution was equilibrated in a thermostatic water bath maintained at ± 0.01 K of the desired temperature, removed from the bath, properly dried, and weighed in an electronic balance. The evaporation losses remained insignificant during the time of actual measurements. Averages of triplicate measurements were taken.

The mixtures were prepared by mixing known masses of pure liquids in airtight stoppered bottles. The reproducibility in mole fraction was within ± 0.0002. The mass measurements, accurate to ± 0.01 mg, were made on a digital electronic analytical

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Table 2. Values of Density ρ, Viscosity η, Excess Molar Volume V^E, Viscosity Deviation $\Delta\eta$, and Grunberg and Nissan Interaction Parameters d_{12} for Binary Mixtures

x_1	$\rho\cdot10^{-3}$ $\text{kg}\cdot\text{m}^{-3}$	η $\text{mPa}\cdot\text{s}$	$V^E\cdot10^6$ $\text{m}^3\cdot\text{mol}^{-1}$	$\Delta\eta$ $\text{mPa}\cdot\text{s}$	d_{12}	x_1	$\rho\cdot10^{-3}$ $\text{kg}\cdot\text{m}^{-3}$	η $\text{mPa}\cdot\text{s}$	$V^E\cdot10^6$ $\text{m}^3\cdot\text{mol}^{-1}$	$\Delta\eta$ $\text{mPa}\cdot\text{s}$	d_{12}
Acrylonitrile + Cinnamaldehyde (298.15 K)											
0	1.046	3.478	0	0	0	0.7889	0.886	0.560	−0.654	−0.451	−0.069
0.2168	1.017	2.014	−0.329	−0.786	−0.267	0.8532	0.862	0.474	−0.526	−0.336	−0.253
0.3837	0.990	1.400	−0.644	−0.878	−0.103	0.9088	0.839	0.423	−0.408	−0.213	−0.239
0.5163	0.964	1.020	−0.831	−0.843	−0.142	0.9573	0.817	0.388	−0.260	−0.097	0.094
0.6241	0.937	0.795	−0.816	−0.731	−0.160	1	0.794	0.344	0	0	0
0.7135	0.911	0.646	−0.752	−0.600	−0.194						
Acrylonitrile + Cinnamaldehyde (308.15 K)											
	1.039	2.688	0	0	0	0.7889	0.881	0.519	−0.749	−0.314	0.119
0.2168	1.011	1.698	−0.504	−0.481	0.039	0.8532	0.856	0.441	−0.614	−0.241	−0.135
0.3837	0.985	1.215	−0.837	−0.571	0.122	0.9088	0.833	0.405	−0.439	−0.147	0.094
0.5163	0.958	0.906	−0.947	−0.568	0.062	0.9573	0.810	0.359	−0.226	−0.079	−0.503
0.6241	0.931	0.724	−0.923	−0.497	0.065	1	0.788	0.307	0	0	0
0.7135	0.905	0.603	−0.850	−0.407	0.084						
Acrylonitrile + Cinnamaldehyde (318.15 K)											
0	1.032	2.296	0	0	0	0.789	0.878	0.457	−1.075	−0.284	−0.233
0.217	1.009	1.456	−1.026	−0.413	−0.051	0.853	0.854	0.430	−0.897	−0.184	0.183
0.384	0.983	1.078	−1.338	−0.461	0.134	0.909	0.830	0.394	−0.640	−0.111	0.456
0.516	0.957	0.830	−1.449	−0.448	0.144	0.957	0.805	0.348	−0.275	−0.061	−0.138
0.624	0.930	0.684	−1.423	−0.382	0.237	1	0.783	0.284	0	0	0
0.714	0.904	0.581	−1.304	−0.308	0.326						
Acrylonitrile + Anisaldehyde (298.15 K)											
0	1.120	3.706	0	0	0	0.794	0.903	0.545	−0.322	−0.492	−0.181
0.222	1.079	2.035	−0.327	−0.925	−0.418	0.857	0.873	0.415	−0.244	−0.410	−1.242
0.391	1.040	1.341	−0.478	−1.051	−0.368	0.911	0.845	0.435	−0.156	−0.207	0.301
0.524	1.003	0.865	−0.549	−1.080	−0.842	0.958	0.819	0.379	−0.059	−0.104	−0.021
0.631	0.968	0.742	−0.510	−0.842	−0.465	1	0.794	0.344	0	0	0
0.720	0.934	0.550	−0.434	−0.737	−0.981						
Acrylonitrile + Anisaldehyde (308.15 K)											
0	1.112	2.830	0.000	0.000	0.000	0.794	0.898	0.469	−0.511	−0.359	−0.220
0.222	1.072	1.828	−0.387	−0.442	0.322	0.857	0.868	0.404	−0.415	−0.264	−0.363
0.391	1.034	1.226	−0.652	−0.618	0.130	0.911	0.840	0.369	−0.309	−0.162	−0.182
0.524	0.997	0.852	−0.712	−0.657	−0.154	0.958	0.813	0.333	−0.140	−0.080	−0.342
0.631	0.962	0.678	−0.696	−0.560	−0.121	1	0.788	0.307	0	0	0.000
0.720	0.930	0.538	−0.663	−0.477	−0.314						
Acrylonitrile + Anisaldehyde (318.5 K)											
0	1.105	2.232	0	0	0	0.794	0.893	0.443	−0.631	−0.243	0.118
0.222	1.066	1.530	−0.502	−0.270	0.462	0.857	0.864	0.390	−0.525	−0.173	0.183
0.391	1.029	1.095	−0.798	−0.376	0.393	0.911	0.835	0.370	−0.357	−0.087	1.000
0.524	0.992	0.798	−0.840	−0.414	0.207	0.958	0.808	0.322	−0.132	−0.043	0.996
0.631	0.957	0.644	−0.791	−0.359	0.248	1	0.783	0.284	0	0	0
0.720	0.924	0.515	−0.714	−0.316	0.081						
Acrylonitrile + Benzaldehyde (298.5 K)											
0	1.041	1.399	0	0	0	0.750	0.887	0.649	−0.729	0.041	1.512
0.182	1.022	1.224	−0.740	0.017	0.817	0.824	0.863	0.565	−0.605	0.035	1.714
0.333	0.995	1.079	−1.062	0.032	0.937	0.889	0.839	0.484	−0.400	0.022	1.871
0.462	0.969	0.958	−1.182	0.046	1.080	0.947	0.816	0.410	−0.198	0.011	2.061
0.571	0.940	0.842	−1.053	0.046	1.201	1	0.794	0.344	0	0	0
0.667	0.914	0.740	−0.948	0.044	1.341						
Acrylonitrile + Benzaldehyde (308.15 K)											
0	1.034	1.224	0	0	0	0.750	0.880	0.550	−0.772	0.013	1.256
0.182	1.011	1.071	−0.826	0.014	0.792	0.824	0.856	0.478	−0.624	0.009	1.365
0.333	0.985	0.941	−1.134	0.022	0.889	0.889	0.833	0.416	−0.451	0.007	1.509
0.462	0.959	0.827	−1.254	0.026	0.986	0.947	0.810	0.358	−0.286	0.002	1.571
0.571	0.932	0.724	−1.175	0.023	1.077	1	0.788	0.307	0	0	0
0.667	0.906	0.630	−1.022	0.017	1.156						
Acrylonitrile + Benzaldehyde (318.15 K)											
0	1.027	1.120	0	0	0	0.750	0.877	0.500	−1.048	0.007	1.185
0.182	1.005	0.973	−0.914	0.005	0.729	0.824	0.854	0.436	−0.977	0.004	1.284
0.333	0.980	0.851	−1.285	0.009	0.821	0.889	0.830	0.379	−0.740	0.002	1.382
0.462	0.954	0.746	−1.399	0.012	0.915	0.947	0.806	0.328	−0.377	0.000	1.463
0.571	0.928	0.653	−1.366	0.011	1.002	1	0.783	0.284	0	0	0
0.667	0.902	0.571	−1.245	0.009	1.088						

balance (Mettler, AG 285, Switzerland). The total uncertainty of density is ± $3\cdot10^{-4}$ $\text{g}\cdot\text{cm}^{-3}$, and that of temperature is ± 0.01 K.

The viscosity was measured by means of a suspended Ubbelohde-type viscometer, which was calibrated at 298.15 K with triple-distilled water and purified methanol using density and viscosity values from the literature. The flow times were accurate to ± 0.1 s, and the uncertainty in the viscosity measurements, based on our work on several pure liquids, was within 0.03 % of the reported value. Details of the methods

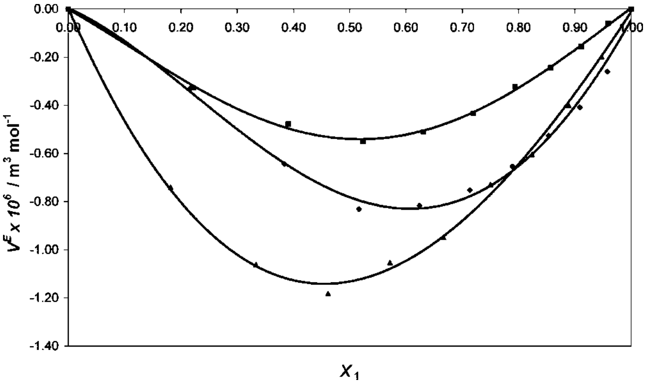

Figure 1. Excess molar volume (V^E) for binary mixtures of acrylonitrile (1) with ♦, cinnamladehyde; ■, anisaldehyde; ▲, benzaldehyde at 298.15 K.

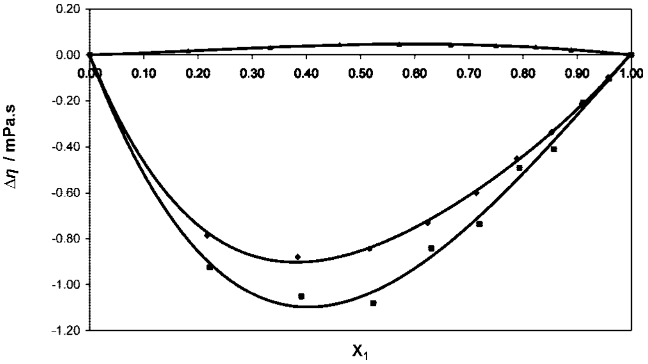

Figure 2. Viscosity deviations ($\Delta\eta$) for binary mixtures of acrylonitrile (1) with ♦, cinnamladehyde; ■, anisaldehyde; ▲, benzaldehyde at 298.15 K.

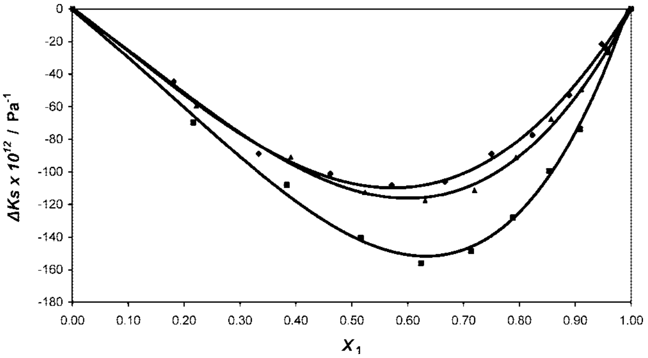

Figure 3. Deviations in isentropic compressibility (ΔK_S) for binary mixtures of acrylonitrile (1) with ♦, cinnamladehyde; ■, anisaldehyde; ▲, benzaldehyde at 298.15 K.

and techniques of density and viscosity measurements have been described earlier.[8–10]

Speeds of sound were determined by a multifrequency ultrasonic interferometer (Mittal Enterprise, New Delhi) working at 5 MHz, calibrated with water, methanol, and benzene at 298.15 K. The details of the methods and techniques have been described earlier.[8,9] The uncertainty of ultrasonic speed measurements is ± 0.2 m·s^{-1}.

Results and Discussion

The experimental viscosities, densities, excess volumes V^E, and viscosity deviations $\Delta\eta$ for the binary mixtures studied at all the experimental temperatures are listed in Table 2. The plots of V^E and $\Delta\eta$ against x_1 at 298.15 K are represented in Figures 1 and 2, respectively. Because of similarity in nature, the plots at the other two temperatures are not presented here.

Table 3. Values of Ultrasonic Speeds *u*, Isentropic Compressibility K_S, and Deviations in Isentropic Compressibility ΔK_S for Binary Mixtures at 298.15 K

x_1	u / m·s^{-1}	$K_S \cdot 10^{12}$ / Pa^{-1}	$\Delta K_S \cdot 10^{12}$ / Pa^{-1}
	Acrylonitrile + Cinnamaldehyde		
0.217	1487.6	444.47	−69.73
0.384	1433.3	491.8	−108
0.516	1403	527.27	−140.5
0.624	1372	567.04	−156
0.714	1330.3	620.18	−148.7
0.789	1288.7	679.46	−128.03
0.853	1251.2	740.98	−99.5
0.909	1224.1	795.28	−73.7
0.957	1186.8	869.05	−24.8
	Acrylonitrile + Anisaldehyde		
0.222	1446.6	442.77	−59.6
0.391	1384.8	501.29	−90.83
0.524	1345.8	550.24	−112.5
0.631	1310	601.97	−117.8
0.720	1277.6	655.57	−111.2
0.794	1244.6	715.18	−91.01
0.857	1217.8	772.21	−67.5
0.911	1201.6	819.38	−49.2
0.959	1186.8	867.06	−26.63
	Acrylonitrile + Benzaldehyde		
0.182	1412.7	490.42	−44.56
0.333	1394.7	516.49	−89
0.462	1353.1	563.95	−101.2
0.571	1322.4	608.09	−108.2
0.667	1292.8	654.64	−105.98
0.750	1259.4	710.4	−89
0.824	1237.6	756.24	−77.37
0.889	1212	811.21	−52.82
0.947	1186.8	869.85	−21.4

The excess molar volume, V^E, was calculated using the equation[11,12]

$$V^E = \sum_{i=1}^{j} x_i M_i (1/\rho - 1/\rho_i) \qquad (1)$$

where ρ is the density of the mixture and M_i, x_i, and ρ_i, are the molecular weight, mole fraction, and density of the *i*th component in the mixture, respectively.

The deviation in viscosities, $\Delta\eta$, was computed using the relationship[11]

$$\Delta\eta = \eta - \sum_{i=1}^{j} (x_i \eta_i) \qquad (2)$$

where η is the dynamic viscosity of the mixture and x_i and η_i are the mole fraction and viscosity of the *i*th component in the mixture, respectively.

It is seen that the values of V^E and $\Delta\eta$ (see Table 2) for all the experimental binary mixtures are negative except benzaldehyde, over the entire range of compositions and temperatures. The negative values of V^E decrease for the three systems in the following order: acrylonitrile + benzaldehyde > acrylonitrile + cinnamaldehyde > acrylonitrile + anisaldehyde.

The negative values of V^E suggest specific interactions[13] between the mixing components resulting in a volume decrease. The negative values of V^E may be attributed to the dipole-induced dipole interactions between the component liquids of the mixtures resulting in formation of electron donor−acceptor complexes.[14] Since the molar volume values of acrylonitrile and the other components differ considerably, nonassociated acry-

Table 4. Redlich−Kister Coefficients a_k and Standard Deviations σ for the Binary Mixtures

excess property	T/K	a_0	a_1	a_2	a_3	a_4	a_5	a_6	σ
			Acrylonitrile + Cinnamaldehyde						
$V^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−3.085	−1.746						0.003
	308.15	−3.747	−0.722	0.109	−1.854				0.005
	318.15	−5.769	−0.580	−1.710					0.024
$\Delta\eta/(mPa \cdot s)$	298.15	−3.418	1.181	0.005	0.849	−2.748	1.816		0.005
	308.15	−2.270	0.765	−0.298					0.007
	318.15	−1.776	0.637	−0.667	0.435				0.014
$\Delta K_S \cdot 10^{12}/(Pa^{-1})$	298.15	−565.107	−341.903						5.438
			Acrylonitrile + Anisaldehyde						
$V^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−2.163	−0.713	1.137	4.926	−4.841	−9.772	10.514	0.008
	308.15	−2.011	0.851						0.018
	318.15	−3.379	0.141	0.250	−4.753	1.141	4.291	−4.99	0.003
$\Delta\eta/(mPa \cdot s)$	298.15	−4.237	1.730						0.035
	308.15	−2.579	−0.276	0.539					0.009
	318.15	−2.579	−0.276	0.539					0.009
$\Delta K_S \cdot 10^{12}/(Pa^{-1})$	298.15	−443.057	−202.522						2.753
			Acrylonitrile + Benzaldehyde						
$V^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−4.524	0.823						0.024
	308.15	−4.894	0.004						0.038
	318.15	−5.536	0.738	−1.773	−2.524				0.034
$\Delta\eta/(mPa \cdot s)$	298.15	0.177	0.834						0.002
	308.15	0.098	−0.041	−0.053	0.049				0.001
	318.15	0.047	0.002	0.041					0.000
$\Delta K_S \cdot 10^{12}/(Pa^{-1})$	298.15	−428.482	−144.086	41.169					3.539

lonitrile molecules are interstitially accommodated into the cluster of bulky aldehyde molecules yielding negative V^E values. This implies the negative V^E values for all the three binary mixtures. As far as $\Delta\eta$ values are concerned, the experimental binary mixture comprised of acrylonitrile and benzaldehyde exhibits positive values, while the other two binary systems exhibit negative values. The negative values imply the presence of dispersion forces[11] between the mixing components in these mixtures, while positive values may be attributed to the presence of specific interactions[11] between them.

Isentropic compressibilities, K_S, and deviations in isentropic compressibility, ΔK_S, are calculated from the experimental densities, ρ, and speeds of sound, u, using the following equations[15–18]

$$K_S = 1/u^2\rho \tag{4}$$

$$\Delta K_S = K_S - \sum_{i=1}^{j} x_i K_{S,i} \tag{5}$$

where K_S is the isentropic compressibility of the mixture and x_i and $K_{S,i}$ are the mole fraction and isentropic compressibility of the ith component in the mixture, respectively. Experimental values of u, K_S, and ΔK_S are compiled in Table 3, and the plots of ΔK_S against x_1 are shown in Figure 3.

It is seen from the ΔK_S vs x_1 curve that the deviations in isentropic compressibility are negative for the three investigated binary mixtures and decrease in the following order: acrylonitrile + cinnamaldehyde > acrylonitrile + anisaldehyde > acrylonitrile + benzaldehyde.

These results can be qualitatively explained by considering the following factors: (i) the mutual disruption of associates present in pure liquids, (ii) dipole-induced interaction between the mixing liquids, (iii) interstitial accommodation of one component into another. The second two factors contribute to negative ΔK_S values. The observed negative value of ΔK_S for the mixtures over the entire range of compositions implies a weak dipole. This induced interaction is predominant between the unlike molecules along with interstitial accommodation between the components.[19,11] Thus, the behaviors of these functions support the results obtained earlier.

Correlating Equations. Grunberg and Nissan[20] suggested the following logarithmic relation between the viscosity of the binary mixture and the pure components

$$\ln \eta_{mix} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d_{12} \tag{6}$$

where d_{12} is a constant proportional to the interchange energy. It may be regarded as an approximate measure of the strength of the molecular interactions between the mixing components. The values of the interchange parameter d_{12} have been calculated using eq 6 as a function of the composition of the binary mixtures of acrylonitrile with cinnamaldehyde, *p*-anisaldehyde, and benzaldehyde and are listed in Table 2.

The mixing functions V^E and $\Delta\eta$ and ΔK_S were fitted by a Redlich−Kister polynomial equation[21]

$$Y^E = x_1 x_2 \sum_{k=1}^{m} a_k (x_1 - x_2)^k \tag{7}$$

where Y^E refers to excess properties; x_1 and x_2 are the mole fraction of acrylonitrile and the other component, respectively; and a_k represents the coefficients. The coefficients a_k were obtained by fitting eq 7 to experimental results using a least-squares regression method. In each case, the optimal number of coefficients was ascertained from an approximation of the variation in the standard deviation (δ). The calculated values of a_k along with the tabulated standard deviation (δ) are listed in Table 4. The standard deviations (δ) was calculated using

$$\delta = [(Y^E{}_{exptl} - Y^E{}_{calcd})^2/(n - m)]^{1/2} \tag{8}$$

where n represents the number of data points and m is the number of coefficients. The small values of σ for excess properties indicate the fits are good for the present study. In the present study, V^E, $\Delta\eta$, and ΔK_S are quite systematic and a function of the composition of the binary mixtures.

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