Volumetric and Transport Properties of Binary Liquid Mixtures of Phenylacetonitrile with Aliphatic Esters at Temperatures of (303.15 to 313.15) K

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The present paper reports the experimental data for density, ρ, viscosity, η, and speed of sound, u, for the binary mixtures of phenylacetonitrile (PAN) with aliphatic esters [ethyl acetate (EA), ethyl chloroacetate (ECA), and ethyl cyanoacetate (ECNA)] over the miscibility region ($0 < x_1 < 1$) at temperatures of (303.15, 308.15, and 313.15) K. The experimental data have been used to calculate various properties like the excess molar volume V^E and deviation in isentropic compressibility $\Delta\kappa_s$. These properties are used to interpret the molecular interactions in the binary mixtures. The Redlich−Kister polynomial equation was fitted to the experimental data to derive binary coefficients and standard deviations.

1. Introduction

Thermodynamic behavior of mixtures containing phenylacetonitrile (PAN) and aliphatic esters are of considerable interest because of their industrial importance. PAN and esters are good industrial solvents having various applications in the synthesis of dyes, pesticides, pharmaceuticals, insecticides and agrochemicals. Thus, correlation of the molecular structures with thermodynamic properties of such mixtures is necessary to obtain systematic information on the behavior of components in the binary mixtures of (PAN + ethyl acetate (EA)), (PAN + ethyl chloroacetate (ECA)), and (PAN + ethyl cyanoacetate (ECNA)). It is easy to understand the importance of the availability of the solvent thermodynamic parameters, such as density, ρ, viscosity, η, and speed of sound, u, which are three important physical properties of solvent systems and are often used to explain the medium effects of solvent on transport phenomena, electrolyte behavior, and reaction mechanisms in solutions.[1] As is well-known, these properties are functionally dependent on the temperature and at least for binary mixtures on the composition of the solvent systems.[2]

To the best of our knowledge, for the mixtures of PAN with aliphatic esters (+ EA, + ECA, and + ECNA) studied in this paper, no experimental data on density, viscosity, and speed of sound as a function of temperature are available in the accessible literature.

The aim of the present work is to analyze the changes in the thermodynamic properties, as a function of temperature and composition of the mixture. For that purpose, density, ρ, viscosity, η, and speed of sound, u, are measured within the temperature range from (303.15 to 313.15) K. The measured values are used to calculate the various properties such as excess molar volumes V^E and deviations in isentropic compressibility $\Delta\kappa_s$.

As a continuation of our research work,[3−8] now we report the experimental data for density, ρ, viscosity, η, and speed of sound, u, for the binary mixtures of PAN with aliphatic esters (+ EA, + ECA, and + ECNA) over the entire range of composition at the temperatures of (303.15, 308.15, and 313.15) K. The results are enlightened in terms of molecular interactions present in the mixtures.

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Table 1. Experimental Density (ρ), Viscosity (η), and Speed of Sound (u) of Pure Liquids at T = 303.15 K

	$10^{-3}\cdot\rho/(\text{kg}\cdot\text{m}^{-3})$		$10^{3}\cdot\eta/(\text{mPa}\cdot\text{s})$		$u/(\text{m}\cdot\text{s}^{-1})$	
component	exptl	lit.	exptl	lit.	exptl	lit.
PAN	1.0088		1.761		1519.2	
EA	0.8889	0.8885[9]	0.379	0.403[10]	1118.5	1118[9]
ECA	1.1393	1.1393[11]	0.969	1.007[11]	1249.0	1249[11]
ECNA	1.0564[a]	1.0564[12]	2.501[a]	2.500[12]	1396.9	

[a] At T = 298.15 K.

2. Experimental Section

2.1. Materials. All of the component liquids are of analytical grade. The PAN and EA were procured from SD Fine Chemicals, India; ECA and ECNA were supplied by Aldrich Chemicals, U.S. The stated mass fraction purities of these are as follows: PAN (0.99), EA (0.995), ECA (0.99), and ECNA (0.98). These solvents were used without further purification. The densities, viscosities, and speeds of sound of pure substances and their comparison with literature values are shown in Table 1.[9−12]

2.2. Apparatus and Procedure. Binary mixtures were prepared by mass in airtight bottles. The mass measurements were performed on a Dhona 100 DS, India, single-pan analytical balance with a resolution of $0.01\cdot10^{-6}$ kg. The required properties of the mixture were measured on the same day of preparing the mixtures. The uncertainty in mole fraction was estimated to be less than $\pm\ 1\cdot10^{-4}$.

Densities of pure liquids and their mixtures were determined by using a $1\cdot10^{-5}\ \text{m}^3$ double arm pycnometer.[13] The uncertainties in density and excess molar volume values were found to be $\pm\ 4\cdot10^{-5}\ \text{g}\cdot\text{cm}^{-3}$ and $\pm\ 1\cdot10^{-3}\ \text{cm}^3\cdot\text{mol}^{-1}$, respectively.

A suspended level viscometer[14,15] was used to measure the flow times of pure liquids and liquid mixtures, and it was calibrated with benzene and doubly distilled water [water conductivity less than $1\cdot10^{-6}\ \Omega^{-1}\cdot\text{cm}^{-1}$ with (0.9970 and 0.9940) $\text{g}\cdot\text{cm}^{-3}$ as its density at (298.15 and 308.15) K,

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respectively, and the density of benzene (0.87381 and 0.87341) $g \cdot cm^{-3}$ at (298.15 and 308.15) K, respectively]. Viscosity values (η) of pure liquids and mixtures were calculated using the flow times by relation

$$\eta = (at - b/t)\rho \qquad (1)$$

where a and b are the characteristic constants of the viscometer, ρ is the density, and t represents the flow time. The flow times of pure liquids and liquid mixtures were determined by the

Table 2. Values of Density (ρ), Viscosity (η), Speed of Sound (U), Excess Molar Volume (V^E), and Deviation in Isentropic Compressibility ($\Delta\kappa_s$) for All Binary Liquid Mixtures at Various Temperatures

x_1	$10^{-3}\cdot\rho$/ ($kg\cdot m^{-3}$)	$10^3\cdot\eta$/ (mPa·s)	u/ ($m\cdot s^{-1}$)	$10^6\cdot V^E$/ ($m^3\cdot mol^{-1}$)	$10^{11}\cdot\Delta\kappa_s$/ ($m^2\cdot N^{-1}$)	x_1	$10^{-3}\cdot\rho$/ ($kg\cdot m^{-3}$)	$10^3\cdot\eta$/ (mPa·s)	u/ ($m\cdot s^{-1}$)	$10^6\cdot V^E$/ ($m^3\cdot mol^{-1}$)	$10^{11}\cdot\Delta\kappa_s$/ ($m^2\cdot N^{-1}$)
					PAN (1) + EA (2)						
					T = 303.15 K						
0.0000	0.8889	0.379	1118.5	0.0000	0.000	0.4610	0.9553	0.839	1326.1	–0.7138	–6.887
0.0198	0.8924	0.396	1125.1	–0.0800	–0.314	0.5665	0.9675	0.984	1366.7	–0.6807	–6.173
0.1279	0.9104	0.483	1173.8	–0.4379	–3.316	0.6978	0.9815	1.180	1417.8	–0.5698	–4.942
0.2425	0.9268	0.591	1230.8	–0.5736	–5.883	0.8328	0.9949	1.416	1464.3	–0.4151	–2.946
0.3589	0.9423	0.714	1286.7	–0.6553	–7.216	0.9745	1.0065	1.695	1508.3	–0.0380	–0.304
						1.0000	1.0088	1.761	1519.2	0.0000	0.000
					T = 308.15 K						
0.0000	0.8826	0.361	1097.8	0.0000	0.000	0.4610	0.9509	0.781	1312.5	–0.8501	–7.717
0.0198	0.8874	0.376	1109.3	–0.2232	–1.270	0.5665	0.9633	0.914	1355.1	–0.8272	–6.963
0.1279	0.9054	0.458	1164.8	–0.5646	–5.217	0.6978	0.9781	1.092	1410.8	–0.7781	–5.797
0.2425	0.9227	0.556	1218.2	–0.7860	–7.230	0.8328	0.9918	1.307	1461.3	–0.6316	–3.698
0.3589	0.9380	0.668	1278.4	–0.8292	–8.807	0.9745	1.0026	1.554	1500.5	–0.1381	–0.319
						1.0000	1.0041	1.602	1512.9	0.0000	0.000
					T = 313.15 K						
0.0000	0.8759	0.344	1081.2	0.0000	0.000	0.4610	0.9465	0.733	1303.1	–1.0243	–8.663
0.0198	0.8808	0.357	1104.3	–0.2301	–3.331	0.5665	0.9585	0.854	1344.9	–0.9202	–7.742
0.1279	0.8996	0.431	1159.7	–0.6502	–7.220	0.6978	0.9724	1.017	1400.0	–0.7391	–6.303
0.2425	0.9169	0.524	1209.3	–0.8440	–8.577	0.8328	0.9852	1.209	1446.8	–0.4557	–3.626
0.3589	0.9334	0.627	1263.6	–0.9890	–9.481	0.9745	0.9981	1.430	1490.0	–0.1526	–0.314
						1.0000	0.9995	1.489	1503.4	0.0000	0.000
					PAN (1) + ECA (2)						
					T = 303.15 K						
0.0000	1.1393	0.969	1229.4	0.0000	0.000	0.4843	1.0726	1.336	1370.5	0.1059	–0.822
0.0205	1.1358	0.982	1235.7	0.0586	–0.079	0.5875	1.0593	1.419	1400.6	0.1005	–0.786
0.1376	1.1192	1.069	1268.8	0.0902	–0.349	0.7152	1.0432	1.524	1438.2	0.0858	–0.680
0.2585	1.1026	1.161	1303.2	0.1019	–0.535	0.8434	1.0275	1.628	1476.9	0.0533	–0.550
0.3793	1.0864	1.254	1338.7	0.1082	–0.698	0.9764	1.0116	1.740	1516.0	0.0134	–0.267
						1.0000	1.0088	1.761	1519.2	0.0000	0.000
					T = 308.15 K						
0.0000	1.1325	0.898	1212.4	0.0000	0.000	0.5875	1.0564	1.302	1388.9	–0.1801	–0.972
0.0205	1.1298	0.912	1218.9	–0.0128	–0.132	0.7152	1.0400	1.394	1426.3	–0.1415	–0.713
0.1376	1.1143	0.991	1254.0	–0.0605	–0.570	0.8434	1.0237	1.488	1466.0	–0.0785	–0.490
0.2585	1.0985	1.072	1291.5	–0.1101	–0.971	0.9764	1.0070	1.584	1505.6	–0.0075	–0.066
0.3793	1.0833	1.156	1328.4	–0.1838	–1.183	1.0000	1.0041	1.602	1512.9	0.0000	0.000
					T = 313.15 K						
0.0000	1.1259	0.834	1197.6	0.0000	0.000	0.4843	1.0628	1.139	1348.5	–0.0536	–1.362
0.0205	1.1232	0.846	1204.0	–0.0045	–0.078	0.5875	1.0498	1.207	1381.0	–0.0492	–1.291
0.1376	1.1076	0.916	1240.4	–0.0185	–0.654	0.7152	1.0340	1.292	1419.9	–0.0412	–1.110
0.2585	1.0917	0.993	1279.0	–0.0331	–1.112	0.8434	1.0183	1.379	1457.4	–0.0305	–0.636
0.3793	1.0762	1.070	1316.6	–0.0472	–1.327	0.9764	1.0023	1.471	1495.8	–0.0052	–0.040
						1.0000	0.9995	1.489	1503.4	0.0000	0.000
					PAN (1) + ECNA (2)						
					T = 303.15 K						
0.0000	1.0514	2.186	1396.9	0.0000	0.000	0.4842	1.0289	1.995	1448.0	0.1222	0.530
0.0223	1.0501	2.179	1399.6	0.0285	0.011	0.5882	1.0246	1.950	1461.3	0.1109	0.479
0.1383	1.0444	2.135	1412.3	0.0775	0.119	0.7172	1.0194	1.893	1479.4	0.0916	0.322
0.2569	1.0388	2.088	1424.4	0.1069	0.278	0.8426	1.0146	1.835	1496.9	0.0634	0.185
0.3778	1.0334	2.040	1436.0	0.1197	0.477	0.9759	1.0097	1.773	1515.5	0.0152	0.045
						1.0000	1.0088	1.761	1519.2	0.0000	0.000
					T = 308.15 K						
0.0000	1.0465	1.962	1384.8	0.0000	0.000	0.4842	1.0263	1.799	1436.3	–0.1268	0.581
0.0233	1.0456	1.955	1386.5	–0.0170	0.073	0.5882	1.0218	1.761	1450.4	–0.1139	0.524
0.1383	1.0409	1.917	1397.7	–0.0652	0.282	0.7172	1.0162	1.713	1468.5	–0.0860	0.430
0.2569	1.0359	1.877	1410.0	–0.1024	0.442	0.8426	1.0109	1.664	1487.6	–0.0598	0.259
0.3778	1.0308	1.836	1423.7	–0.1184	0.533	0.9759	1.0052	1.612	1508.7	–0.0222	0.052
						1.0000	1.0041	1.602	1512.9	0.0000	0.000
					T = 313.15 K						
0.0000	1.0408	1.770	1367.7	0.0000	0.000	0.4842	1.0211	1.644	1429.2	–0.1168	0.138
0.0223	1.0398	1.764	1370.4	–0.0023	0.026	0.5882	1.0168	1.612	1442.0	–0.1146	0.126
0.1383	1.0348	1.733	1384.6	–0.0124	0.079	0.7172	1.0114	1.572	1459.4	–0.0973	0.099
0.2569	1.0299	1.701	1399.1	–0.0379	0.109	0.8426	1.0058	1.535	1478.0	–0.0253	0.049
0.3778	1.0253	1.671	1414.2	–0.0961	0.128	0.9759	1.0004	1.496	1495.9	–0.0029	0.009
						1.0000	0.9995	1.489	1503.4	0.0000	0.000

average of five measurements. The uncertainty of viscosity was found to be ± 0.005 mPa·s.

Speeds of sound was determined by using an ultrasonic interferometer (model M-82, Mittal Enterprises, India), working at a 2 MHz frequency. The working principle used in the measurement of the speed of sound through a medium was based on the accurate determination of the wavelength of ultrasonic waves of known frequency produced by a quartz crystal in the measuring cell.[3,16] The temperature of the solution was controlled by circulating water at a desired temperature through the jacket of the double-walled cell. The speed of sound was measured with relative uncertainty of 0.3 %.

In all of the property measurements, the temperature was controlled within ± 0.01 K using a constant temperature bath (INSREF model IRI-016 C, India), and the temperature was monitored with a platinum resistance thermometer with an accuracy of ± 0.001 K and an uncertainty of ± 0.004 K.

3. Results and Discussion

Table 2 reports the experimental data of density, ρ, viscosity, ρ, ultrasonic speed, u, excess molar volume, V^E, and deviation in isentropic compressibility, $\Delta\kappa_s$, for the binary mixtures of (PAN + EA), (PAN + ECA), and (PAN + ECNA) at T = (303.15, 308.15, and 313.15) K along with the mole fraction.

The excess molar volumes (V^E) have been evaluated from density using

$$V^E/(\mathrm{cm^3\cdot mol^{-1}}) = (x_1M_1 + x_2M_2)/\rho_m - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (2)$$

where ρ_m is the density of the mixture; x_1, M_1, and ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, molar mass, and density of pure components, respectively.

The deviations in isentropic compressibility ($\Delta\kappa_s$) have been evaluated using the equation

$$\Delta\kappa_s/(\mathrm{m^2\cdot N^{-1}}) = \kappa_s - (\Phi_1\kappa_{s1} + \Phi_2\kappa_{s2}) \quad (3)$$

where Φ_i is the volume fraction of pure components and is calculated from the individual pure molar volumes, V_i, using the relation

$$\Phi_i = x_iV_i/(\sum x_iV_i) \quad (4)$$

and κ_{s1}, κ_{s2}, and κ_s are the isentropic compressibility of the pure components and observed isentropic compressibility of the liquid mixture, respectively.

The excess or deviation properties ΔY are fitted by the method of nonlinear least-squares to the fourth-order Redlich–Kister type polynomial equation.[17]

$$\Delta Y = x_1x_2\sum A_i(x_1 - x_2)^i \quad (5)$$

where A_0, A_1, A_2, A_3, and A_4 are adjustable binary coefficients. The coefficients A_i were estimated using multiparametric regression analysis based on a nonlinear least-squares method. The number of A_i parameters was optimized using the F-test and is found to be five (m = 5). In each case, the optimum number of coefficients A_i is determined from an examination of the variation of standard deviation (σ) as calculated by

$$\sigma(Y^E) = [\sum(\Delta Y_{obs} - \Delta Y_{cal})^2/(n - m)]^{1/2} \quad (6)$$

where n represents the number of experimental points and m is the number of coefficients used in fitting the data. The coefficients A_i and standard deviations (σ) V^E, (σ) $\Delta\eta$, and (σ) $\Delta\kappa_s$ of the fit are summarized in Table 3.

3.1. Excess Molar Volume (V^E). Figure 1 depicts the graphical representation of excess molar volumes (V^E) for PAN with EA, ECA, and ECNA at 303.15 K. The strength of unlike molecular interactions in the solution is better estimated by the sign and magnitude of V^E. The values of V^E are negative for the PAN + EA system, whereas they are positive for PAN + ECA and PAN + ECNA systems. The positive contributions are thought to be arising from the disruption of dipolar association of PAN. However, it is observed that the EA show considerable larger negative V^E values compared to other esters. This observation supports the interstitial accommodation of EA molecules due to the smaller size in dipolar network of the PAN.[6,16] The fact that V^E values become more negative with the temperature further supports the interstitial accommodation of EA molecules (Figure 4). The observed negative V^E values for ECA and ECNA systems (Figures 2, 3, and 4) at higher temperatures show the existence of weaker dipolar interaction between unlike molecules.

Table 3. Binary Coefficients A_i and Corresponding Standard Deviations (σ) of Equation 6

property	T/K	A_0	A_1	A_2	A_3	A_4	σ
			PAN (1) + EA (2)				
$10^6\cdot V^E/(\mathrm{m^3\cdot mol^{-1}})$	303.15	−2.780	0.07	−0.70	1.1	−0.4	0.022
	308.15	−3.39	0.08	−1.9	1	−2.2	0.037
	313.15	−4.048	1.08	2.35	0.86	−6.9	0.028
$10^{11}\cdot\Delta\kappa_s/(\mathrm{m^2\cdot N^{1-}})$	303.15	−27.73	9	−2.9	−13.1	17.6	0.1
	308.15	−31.84	11.1	−8	−3.9	2	0.35
	313.15	−35.7	7.4	4	32	−45	0.57
			PAN (1) + ECA (2)				
$10^6\cdot V^E/(\mathrm{m^3\cdot mol^{-1}})$	303.15	0.441	0.02	−0.26	−0.60	1.23	0.012
	308.15	−0.80	−0.03	0.82	−0.05	−0.61	0.0069
	313.15	−0.2101	−0.012	0.130	−0.073	−0.187	0.0014
$10^{11}\cdot\Delta\kappa_s/(\mathrm{m^2\cdot N^{1-}})$	303.15	−3.26	−0.44	3.0	−2	−7.9	0.039
	308.15	−4.94	1.87	4.6	−2.4	−6.0	0.058
	313.15	−5.41	0.01	−2.08	0.27	5.30	0.014
			PAN (1) + ECNA (2)				
$10^6\cdot V^E/(\mathrm{m^3\cdot mol^{-1}})$	303.15	0.4857	−0.093	−0.08	−0.069	0.53	0.0035
	308.15	−0.5005	0.149	0.291	−0.193	−0.65	0.0028
	313.15	−0.48	−0.272	0.76	0.38	−0.2	0.0071
$10^{11}\cdot\Delta\kappa_s/(\mathrm{m^2\cdot N^{-1}})$	303.15	2.130	0.089	−3.37	0.65	2.91	0.0067
	308.15	2.283	0.003	−0.73	−0.29	1.10	0.0094
	313.15	0.5478	0.02	−0.216	−0.391	0.35	0.014

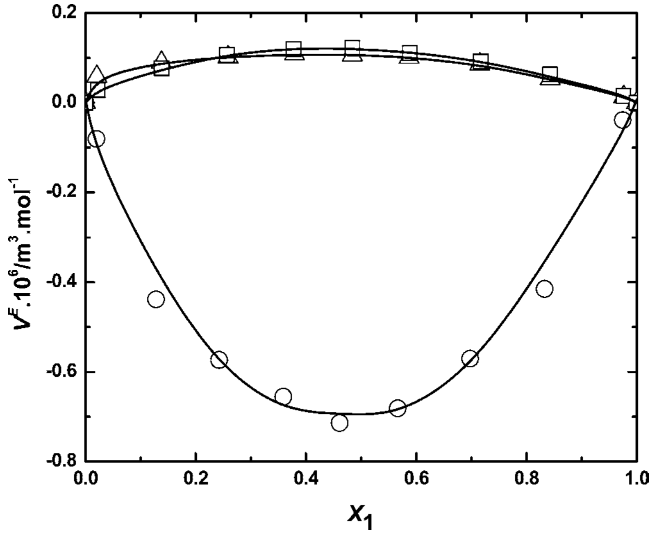

Figure 1. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 303.15 K: ○, {PAN (1) + EA (2)}; △, {PAN (1) + ECA (2)}; □, {PAN (1) + ECNA (2)}; the symbols represent experimental values and solid curves represent the smoothed data of this work.

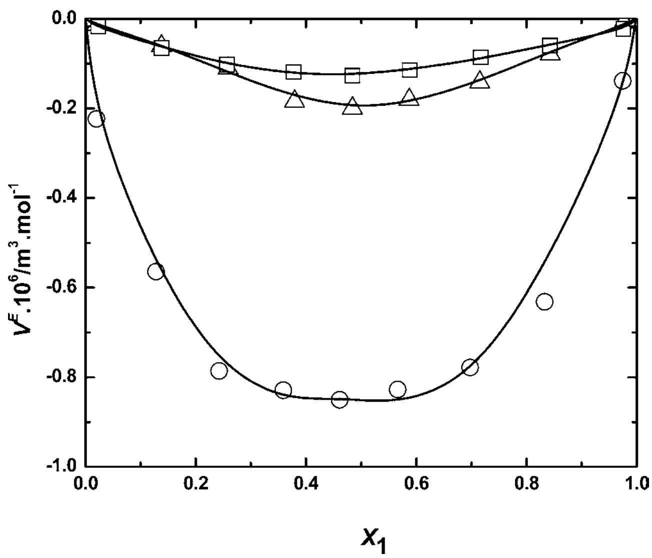

Figure 2. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 308.15 K: ○, {PAN (1) + EA (2)}; △, {PAN (1) + ECA (2)}; □, {PAN (1) + ECNA (2)}; the symbols represent experimental values and solid curves represent the smoothed data of this work.

The algebraic values of V^E of PAN with aliphatic esters fall in the order at 303.15 K,

$$\text{EA} < 0 < \text{ECA} < \text{ECNA}$$

whereas at 308.15 and 313.15 K

$$\text{EA} < \text{ECA} < \text{ECNA} < 0$$

3.2. Deviation in Isentropic Compressibility ($\Delta\kappa_s$). Generally V^E and $\Delta\kappa_s$ have the same sign as observed in the case of PAN + EA and + ECA binary mixtures, while the PAN + ECNA system does not follow the general rule. The $\Delta\kappa_s$ value increases become more negative with the temperature, which further fortified the observation about interstitial accommodation of EA molecules.

The values of $\Delta\kappa_s$ may be attributed to the effects of relative strength that influence the free space defined by Jacobson.[18] In the present investigation the negative $\Delta\kappa_s$ values obtained over a range of temperatures studied for all mixtures are attributed

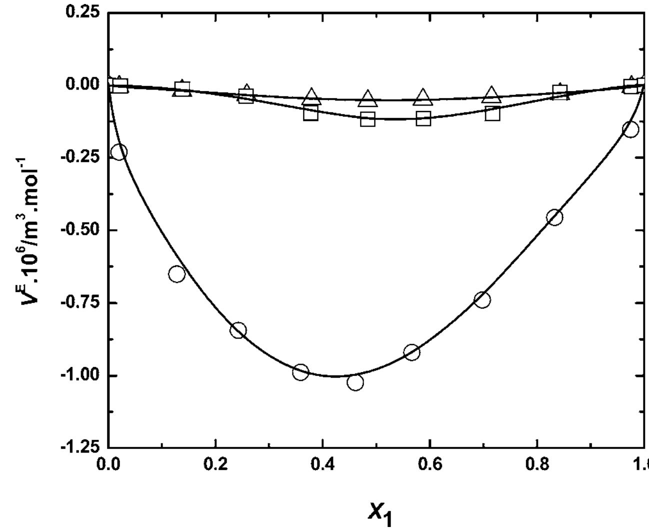

Figure 3. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 313.15 K: ○, {PAN (1) + EA (2)}; △, {PAN (1) + ECA (2)}; □, {PAN (1) + ECNA (2)}; the symbols represent experimental values and solid curves represent the smoothed data of this work.

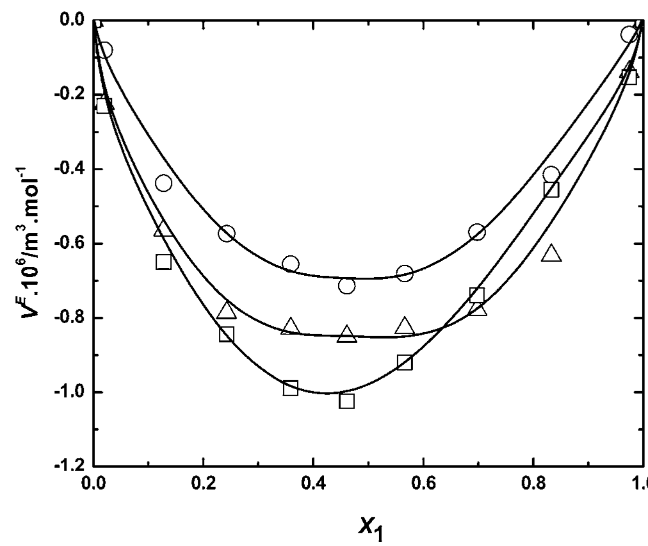

Figure 4. Plots of excess molar volumes, V^E, as a function of mole fraction x of PAN (1) + EA (2) at ○, 303.15; △, 308.15; □, 313.15 K; the symbols represent experimental values and solid curves represent the smoothed data of this work.

to chemical forces operating between unlike molecules of binary mixtures.[19,20] The positive $\Delta\kappa_s$ values for PAN + ECNA system may be attributed to mutual loss of dipolar association.

The algebraic values of $\Delta\kappa_s$ vary in the following order at 303.15 K.

$$\text{EA} < \text{ECA} < 0 < \text{ECNA}$$

4. Conclusion

In this paper, we present the experimental data on density, viscosity, and speeds of sound for three binary mixtures of PAN + EA, ECA, and ECNA at T = (303.15 to 313.15) K. It may also be concluded from the interpretation of derived properties, excess molar volume (V^E) and isentropic compressibility ($\Delta\kappa_s$), that mixing the liquids creates the structure through interstitial accommodation and dipolar−dipolar association in the binary mixtures.

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