

Thermophysical Properties of *para*-Anisaldehyde (1) + Chlorobenzene (2) at Temperatures of (303.15, 313.15, and 323.15) K and a Pressure of 0.1 MPa

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The density, viscosity, refractive index, surface tension and ultrasonic velocity have been determined for (*p*-anisaldehyde + chlorobenzene) over the entire composition range at temperatures of (303.15, 313.15, and 323.15) K. The viscosity values were fit to the models of McAllister, Grundberg and Nissan, Hind, Krishnan and Laddha and Jouyban–Acree. The ultrasonic velocity values were fit to the Jouyban–Acree model and Vandael and Vangeel equation. The excess values were correlated using the Redlich–Kister polynomial equation to obtain their coefficients and standard deviations.

Introduction

Binary liquid mixtures due to their unusual behavior have attracted considerable attention.¹ In chemical process industries, materials are normally handled in fluid form, and as a consequence, the physical, chemical, and transport properties of fluids assume importance. Thus, data on some of the properties associated with the liquids and liquid mixtures like density, viscosity, refractive index, surface tension, and ultrasonic velocities find extensive application in solution theory and molecular dynamics.² Such results are necessary for interpretation of data obtained from thermochemical, electrochemical, biochemical, and kinetic studies.³ The review of literature on acoustical studies of solutions reveals that ultrasonic measurements are used to estimate the different elastic properties. *para*-Anisaldehyde is chemically known as 4-methoxy benzaldehyde and used in perfume and pharmaceutical industries. The *para*-anisaldehyde and chlorobenzene mixture is used as insecticides, as comprising gel formulations for vapor producing systems, and also in the preparation of a polymer containing pendant unsaturation. In our earlier paper,⁴ we had studied the transport properties of binary mixtures composed of 1–4 dioxane, butanol, and carbon tetrachloride. In continuation of this research, we have reported density, viscosity, refractive index, surface tension, and ultrasonic velocity of pure *p*-anisaldehyde and chlorobenzene as well as for the binary system constituted by these two chemicals at temperatures of (303.15, 313.15, and 323.15) K. The viscosity values have been fitted to the McAllister,⁵ Grundberg and Nissan model,⁶ Hind model,⁷ Krishnan and Laddha model,⁸ and Jouyban–Acree model.⁹ The ultrasonic velocity values were fitted to the Jouyban–Acree model and Vandael and Vangeel¹⁰ equation. A literature survey showed that no measurements have been previously reported for the mixture studied in this paper.

Experimental Section

Materials. The *p*-anisaldehyde of cited purity of 99.5 % and chlorobenzene with purity of 99.5 % were obtained from Lobo

Table 1. Comparison of Experimental Density and Viscosity of Pure Liquids with Literature Values at 303.15 K and 0.1 MPa

pure liquids	$\rho \cdot 10^{-3}/(\text{kg} \cdot \text{m}^{-3})$		$\eta/(\text{mPa} \cdot \text{s})$	
	lit.	exptl	lit.	exptl
anisaldehyde	1.1190	—	3.2753	—
chlorobenzene	1.1020	1.1000 ^a	0.7105	0.7053 ^b

^a Ref 15. ^b Ref 16.

chemicals and dried over anhydrous calcium chloride and fractionally distilled before use.¹¹ The purity of the solvents, after purification, was ascertained by comparing their densities and viscosities, with the corresponding literature values at 303.15 K (Table 1). Binary mixtures were gravimetrically prepared with a Shimadzu Corporation Japan type BL 2205 electronic balance with an uncertainty of 0.01 g. Care was taken to avoid evaporation and contamination during mixing. The resulting mole fraction uncertainty was estimated to be less than ± 0.0001 . All the measurements described below were performed at least three times, and the results presented are the average.

Apparatus and Procedure. Densities were determined by using a 15 cm³ bicapillary pycnometer described previously¹² and calibrated with deionized double distilled water with a density of 996.0 kg·m⁻³ at a temperature of 303.15 K. The pycnometer was thermostatted in a transparent walled water bath (maintained constant to ± 0.01 K) for 15 min to attain thermal equilibrium, and the liquid level in the two arms was obtained with a traveling microscope which could read to 0.01 mm. The precision of the density measurements was estimated to be ± 0.0003 g·cm⁻³.

The kinematic viscosities were measured with an Ostwald viscometer previously calibrated using water. The time was measured with a precision of 0.01 s, and the uncertainty in the viscosity was estimated to be less than 0.0003 mPa·s. The kinematic viscosity was obtained from the working equation

$$v = at - b/t \quad (1)$$

where the two constants *a* and *b* were obtained by measuring the flow time *t* of benzene. The viscosities of mixtures of *p*-anisaldehyde and chlorobenzene have been correlated with the model proposed by McAllister for a two-component mixture considering three-body interaction

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$$\ln \nu = x^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln(x_1 + x_2 M_2/M_1) + 3x_1^2 x_2 \ln((2 + M_2/M_1)/3) + x_2^3 \ln(M_2/M_1) + 3x_1 x_2^2 \ln((1 + 2M_2/M_1)/3) \quad (2)$$

In eq 2, ν_1 and ν_2 refer to the kinematic viscosity of pure liquids 1 and 2 having mole fractions x_1 and x_2 , respectively. The parameters ν_{12} and ν_{21} represent the interaction parameters obtained by multiple regression analysis, while M_1 and M_2 are the molar mass of the components. Grundberg and Nissan proposed the following equation

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12} \quad (3)$$

where G_{12} is an interaction parameter which is a function of the components 1 and 2 as well as temperature.

Hind suggested an equation for the viscosity of binary liquid mixture as

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1 x_2 H_{12} \quad (4)$$

where H_{12} is an interaction parameter.

The kinematic viscosity was correlated by means of the Krishnan and Laddha model for a two-component mixture, which gives

$$\ln \nu = x_1 \ln \nu_1 + x_2 \ln \nu_2 + x_1 \ln M_1 + x_2 \ln M_2 + \ln(x_1 M_1 + x_2 M_2 - 2.30x_1 x_2 (B + C(x_1 - x_2) \dots)) \quad (5)$$

where B and C are interaction parameters.

Jouyban et. al proposed a model for correlating the viscosity of liquid mixtures at various temperatures

$$\ln y_{m,T} = f_1 \ln y_1 + f_2 \ln y_2 + f_1 f_2 \sum [A_j (f_1 - f_2)^j / T] \quad (6)$$

where $y_{m,T}$, $y_{1,T}$, and $y_{2,T}$ are the viscosity of the mixture and solvents 1 and 2 at temperature T , respectively. A_j is the model constant.

Refractive indices were measured using a thermostatically controlled Abbe refractometer (Atago 3T) with an uncertainty of less than 0.0001. Water was circulated in to the prism of the refractometer by a circulation pump connected to an external thermostatted water bath. The instrument was calibrated by measuring the refractive indices of doubly distilled water and propyl alcohol. The sample mixture was directly injected into the prism assembly of the instrument using a syringe. The solutions were prethermostatted at the temperature of the experience before the experiments to achieve a quick thermal equilibrium.

Surface tension of pure liquids and binary mixtures over the whole composition range was determined using an Interfacial tensiometer (ASTM D.971) with 1 No. 4 cm platinum ring as per IS 6104. All samples were equilibrated to (303.15, 313.15, and 323.15) K under atmospheric pressure. It was calibrated with distilled water. The accuracy of the surface tension measurement was estimated to be 0.03 mN·m⁻¹.

The speed of sound was measured by using a variable path, single crystal interferometer (Mittal Enterprises, New Delhi) at a frequency of 2 MHz. The interferometer was calibrated using toluene. The interferometer cell was filled with the test liquid, and the temperature of the solution was maintained constant within ± 0.01 K by circulation of water from a thermostatically regulated water bath through the water jacketed cell. The uncertainty was estimated to be 1 m·s⁻¹. Theoretical values of ultrasonic velocity are calculated using eq 6 and the Vandael and Vangeel equation.

$$U = [x_1/m_1 U_1^2 + x_2/m_2 U_2^2] (x_1 m_1 + x_2 m_2)^{-1/2} \quad (7)$$

Results and Discussion

Measured values of densities, viscosities, refractive indices, surface tensions, and ultrasonic velocities of (*p*-anisaldehyde + chlorobenzene) at temperatures of (303.15, 313.15, and 323.15) K are listed in Table 2. The density values have been used to calculate excess molar volumes V^E using the following equation

$$V^E = (x_1 M_1 + x_2 M_2)/\rho_m - (x_1 M_1/\rho_1 + x_2 M_2/\rho_2) \quad (8)$$

where x_1 and x_2 refer to the mole fraction of components 1 and 2. ρ_1 , ρ_2 , and ρ_m refer to the density of components 1 and 2 and the density of the mixture, respectively. The uncertainty in the calculation of V^E from density measurements was estimated to be ± 0.0001 .

The viscosity deviations $\Delta\eta$ were calculated from the viscosity values using

$$\Delta\eta = \eta - (x_1 \eta_1 + x_2 \eta_2) \quad (9)$$

where η , η_1 , and η_2 are the viscosity of the mixture and the viscosity of pure components 1 and 2, respectively. The uncertainty in the calculation of $\Delta\eta$ from viscosity measurements was estimated to be ± 0.0001 .

Table 2. Densities ρ , Viscosities η , Refractive Indices n_D , Surface Tensions σ , and Ultrasonic Velocities U for the *p*-Anisaldehyde (1) + Chlorobenzene (2) Mixture at $T = (303.15, 313.15, \text{ and } 323.15)$ K and 0.1 MPa

x_1^a	$\rho \cdot 10^{-3}$	η	n_D	σ	U
	kg·m ⁻³	mPa·s		mN·m ⁻¹	m·s ⁻¹
$T = 303.15$ K					
0.0000	1.1020	0.7105	1.5590	33.60	1220
0.1440	1.1051	0.8478	1.5589	43.12	1339
0.2517	1.1070	1.0227	1.5587	50.12	1383
0.3354	1.1091	1.1681	1.5586	56.00	1418
0.4308	1.1112	1.3340	1.5585	62.50	1459
0.5457	1.1132	1.5661	1.5583	70.14	1505
0.6271	1.1153	1.7714	1.5581	75.82	1537
0.7371	1.1180	2.0864	1.5578	84.11	1580
0.8079	1.1220	2.3533	1.5576	87.59	1607
0.9439	1.1240	2.9476	1.5572	96.09	1662
1.0000	1.1255	3.2753	1.5571	99.08	1684
$T = 313.15$ K					
0.0000	1.0800	0.6914	1.5509	32.30	797
0.1440	1.0851	0.7705	1.5508	41.60	820
0.2517	1.0893	0.9236	1.5507	49.11	924
0.3354	1.0921	1.0472	1.5506	55.33	1004
0.4308	1.0996	1.1777	1.5504	62.12	1097
0.5457	1.1005	1.3575	1.5502	71.23	1207
0.6271	1.1035	1.4969	1.5500	78.22	1283
0.7371	1.1072	1.7623	1.5497	83.11	1385
0.8079	1.1102	1.9856	1.5494	86.41	1452
0.9439	1.1163	2.5963	1.5490	94.66	1577
1.0000	1.1191	2.9695	1.5488	96.11	1631
$T = 323.15$ K					
0.0000	1.0674	0.5543	1.5480	31.26	695
0.1440	1.0735	0.6646	1.5480	40.52	738
0.2517	1.0786	0.8222	1.5479	47.99	758
0.3354	1.0825	0.9386	1.5478	53.34	850
0.4308	1.0863	1.0594	1.5477	60.11	955
0.5457	1.0916	1.1920	1.5475	68.77	1082
0.6271	1.0950	1.2870	1.5475	75.22	1170
0.7371	1.1000	1.4738	1.5474	82.89	1290
0.8079	1.1030	1.6348	1.5470	85.22	1365
0.9439	1.1101	2.1221	1.5466	93.44	1511
1.0000	1.1141	2.4418	1.5463	94.10	1571

^a Mole fraction of *para*-anisaldehyde.

Table 3. Viscosity Deviation $\Delta\eta$, Excess Volume V^E , Surface Tension Deviation $\Delta\sigma$, and Refractive Index Deviation Δn_D of the *p*-Anisaldehyde (1) + Chlorobenzene (2) Mixture at $T = (303.15, 313.15, \text{ and } 323.15) \text{ K}$ and 0.1 MPa

x_1	$\Delta\eta$	V^E	$\Delta\sigma$	Δn_D
	mPa·s	cm ³ ·mol ⁻¹	mN·m ⁻¹	
$T = 303.15 \text{ K}$				
0.0000	0.0000	0.0000	0.00	0.0000
0.1440	0.1927	0.0850	0.07	0.0014
0.2517	0.2990	0.1056	0.11	0.0017
0.3354	0.3721	0.1555	0.43	0.0017
0.4308	0.4553	0.1992	0.69	0.0023
0.5457	0.5232	0.2518	0.76	0.0031
0.6271	0.5304	0.2702	1.13	0.0033
0.7371	0.5025	0.2138	2.24	0.0029
0.8079	0.4205	0.1645	0.99	0.0020
0.9439	-0.1812	0.0402	0.59	0.0013
1.0000	0.0000	0.0000	0.00	0.0000
$T = 313.15 \text{ K}$				
0.0000	0.0000	0.0000	0.00	0.0000
0.1440	0.1556	0.1394	0.11	0.0030
0.2517	0.2596	0.1519	0.73	0.0032
0.3354	0.3358	0.2009	1.62	0.0040
0.4308	0.4330	0.2386	2.33	0.0040
0.5457	0.5275	0.2918	4.10	0.0044
0.6271	0.5824	0.3032	5.90	0.0041
0.7371	0.5796	0.2589	3.77	0.0034
0.8079	0.5253	0.2433	2.25	0.0019
0.9439	0.2393	0.1303	2.12	0.0008
1.0000	0.0000	0.0000	0.00	0.0000
$T = 323.15 \text{ K}$				
0.0000	0.0000	0.0000	0.00	0.0000
0.1440	0.0621	0.1526	0.19	0.0001
0.2517	0.1203	0.2031	0.82	0.0020
0.3354	0.1716	0.2188	1.80	0.0023
0.4308	0.2420	0.2717	2.70	0.0031
0.5457	0.3396	0.3125	4.60	0.0033
0.6271	0.4077	0.3455	6.90	0.0049
0.7371	0.4413	0.3303	5.30	0.0060
0.8079	0.4221	0.3156	3.10	0.0033
0.9439	0.2073	0.1880	2.80	0.0019
1.0000	0.0000	0.0000	0.00	0.0000

The changes of refractive index (Δn_D), from linear additive value of the mole fraction is obtained by

$$\Delta n_D = n_D - (x_1 n_{D1} + x_2 n_{D2}) \quad (10)$$

In the same way, the changes of surface tension ($\Delta\sigma$), from linear additive value of the mole fraction is obtained by

$$\Delta\sigma = \sigma - (x_1\sigma_1 + x_2\sigma_2) \quad (11)$$

The viscosity deviations, excess volume, surface tension deviation, and refractive index deviation of the *p*-anisaldehyde (1) + chlorobenzene (2) mixture at (303.15, 313.15, and 323.15) K are presented in Table 3. The excess molar volumes, deviations of viscosity, deviations of surface tension, and deviations of refractive index were fitted to a Redlich–Kister¹³ equation of the type

$$Y = x_1 x_2 \sum A_i (x_1 - x_2)^i \quad (12)$$

where Y is either V^E , $\Delta\eta$, Δn_D , or $\Delta\sigma$ and n is the degree of polynomial. Coefficients A_i were obtained by fitting eq 12 to experimental results using a least-squares regression method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (S). S was calculated using the relation

$$S(Y) = [\sum (A_{\text{exptl}} - A_{\text{calcd}})^2 / (N - n)]^{1/2} \quad (13)$$

where N is the number of data points and n is the number of coefficients. The calculated values of coefficients (A_i) along with the standard deviation (S) are given in Table 4.

Table 4. Parameters and Standard Deviations S of Equations 12 and 13 for *p*-Anisaldehyde (1) + Chlorobenzene (2) $T = (303.15, 313.15, \text{ and } 323.15) \text{ K}$ and 0.1 MPa

functions	A_0	A_1	A_2	S
$T = 303.15 \text{ K}$				
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	-0.5601	0.8551	0.2435	1.06
$\Delta\eta/\text{mPa} \cdot \text{s}$	1.3416	0.7952	1.3005	1.46
Δn_D	0.0012	0.0011	0.0004	0.00
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	3.8114	6.6245	15.226	1.73
$T = 313.15 \text{ K}$				
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	1.7503	1.2389	0.7614	0.06
$\Delta\eta/\text{mPa} \cdot \text{s}$	1.1381	0.0009	0.6037	1.19
Δn_D	0.0018	0.0006	0.0015	0.00
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	13.8300	19.7828	-45.4713	0.10
$T = 323.15 \text{ K}$				
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	-2.7720	1.5169	1.2053	1.11
$\Delta\eta/\text{mPa} \cdot \text{s}$	-0.9495	0.8709	0.5936	1.00
Δn_D	0.00164	0.0014	0.0035	0.00
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	14.4820	26.8621	61.788	1.27

Table 5. Parameters of the McAllister, Hind, and Grundberg Constants and Standard Deviation for the Kinematic Viscosity of *p*-Anisaldehyde (1) + Chlorobenzene (2) at $T = (303.15, 313.15, \text{ and } 323.15) \text{ K}$ and 0.1 MPa

T/K	McAllister			Hind		Grundberg and Nissan	
	A	B	S	H_{12}	S	G_{12}	S
303.15	1.7392	1.2524	2.01	0.2721	0.06	-1.048	0.02
313.15	1.4838	1.0592	2.63	0.3436	0.02	-0.921	0.03
323.15	0.9901	1.3929	2.85	0.4321	0.00	0.673	0.03

Table 6. Parameters of Krishnan and Laddha, Jouban–Acree Constants, and Standard Deviation for the Viscosity of *p*-Anisaldehyde (1) + Chlorobenzene (2) at $T = (303.15, 313.15, \text{ and } 323.15) \text{ K}$ and 0.1 MPa

T/K	Krishnan and Laddha			Jouban–Acree		
	A	B	S	A	B	S
303.15	0.1858	-0.0660	2.68	-103.84	-38.64	0.94
313.15	0.2525	0.1039	3.10	-242.60	-14.30	0.18
323.15	0.1207	0.2787	3.32	-224.81	-83.01	0.79

Table 7. Parameters and Standard Deviation of the Jouban–Acree Model and Standard Deviation of the Vandael and Vangeel Equation for the Ultrasonic Velocity of *p*-Anisaldehyde (1) + Chlorobenzene (2) at $T = (303.15, 313.15, \text{ and } 323.15) \text{ K}$ and 0.1 MPa

T/K	Jouban–Acree			Vandael and Vangeel
	A	B	S	S
303.15	41.97	-93.41	0.31	0.17
313.15	5.31	86.60	0.18	0.35
323.15	-33.0	122.36	0.20	0.35

Interaction parameters and standard deviations of the McAllister model, Grundberg–Nissan model, and Hind model for the viscosity of anisaldehyde–chlorobenzene at (303.15, 313.15, and 323.15) K are presented in Table 5. Interaction parameters and standard deviations of Krishnan and Laddha and Jouban–Acree model for the viscosity of anisaldehyde (1) + chlorobenzene (2) at (303.15, 313.15, and 323.15) K are presented in Table 6. It is clear that both single parameter models, Grundberg–Nissan and Hind, are suitable for correlating the kinematic viscosities of the binary mixture studied. The Jouban–Acree model is also suitable for correlating the kinematic viscosities of the studied binary mixture when compared with the Krishnan and Laddha and McAllister model. Constants and standard deviations of the Jouban–Acree model and standard deviation of the Vandael and Vangeel equation for the ultrasonic velocity of anisaldehyde (1) + chlorobenzene (2) at (303.15, 313.15, and 323.15) K are presented in Table 7. It can be seen that no molecular interaction is taken into account

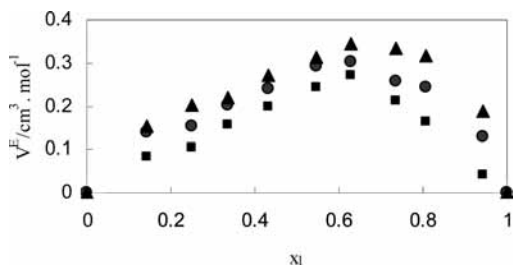


Figure 1. Excess molar volumes (V^E) for [*p*-anisaldehyde (1) + chlorobenzene (2)]: ■, $T = 303.15$ K; ●, $T = 313.15$ K; ▲, $T = 323.15$ K.

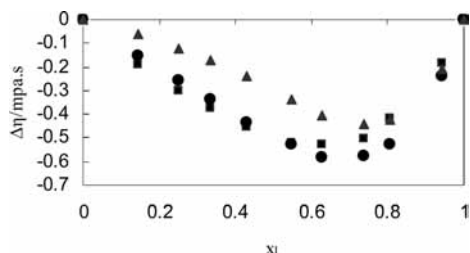


Figure 2. Deviations in viscosity ($\Delta\eta$) for [*p*-anisaldehyde (1) + chlorobenzene (2)]: ■, $T = 303.15$ K; ●, $T = 313.15$ K; ▲, $T = 323.15$ K.

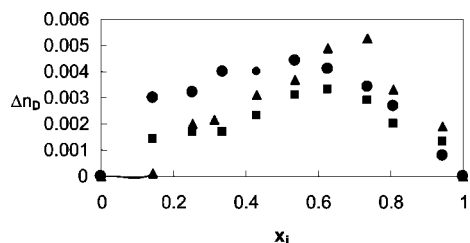


Figure 3. Deviations in refractive index (Δn_D) for [*p*-anisaldehyde (1) + chlorobenzene (2)]: ■, $T = 303.15$ K; ●, $T = 313.15$ K; ▲, $T = 323.15$ K.

for the Vandael and Vangeel equation. The assumption for the formation of ideal mixing relation due to VanDeal and Vangeel is that the ratio of specific heats of the components is equal to the ratio of specific heats of ideal mixtures, and the volumes are also equal. It has been concluded that the Jouban–Acree model provided the best results for ultrasonic velocities of the studied binary mixture when compared with the VanDeal and Vangeel equation which is indicated by low standard deviation values.

The variation of V^E with the mole fraction x_i of anisaldehyde (1) + chlorobenzene (2) at (303.15, 313.15, and 323.15) K is represented in Figure 1. This shows that the excess molar volumes are always positive for all the studied temperatures and for any composition. Figure 2 depicts the variation of $\Delta\eta$ with the mole fraction x_i of *p*-anisaldehyde. $\Delta\eta$ values are always negative for all the studied temperatures and for any composition. The increase in temperature and the viscosity deviation values become less negative. No published work was found for the present system with which to compare our results. However the same trend has been observed by Hianni et. al¹⁴ for the excess volumes and viscosity deviations of several other aldehydes.

The variation of Δn_D with composition of mixture (x_i of *p*-anisaldehyde) is presented in Figure 3. A detailed observation of the figure shows that the values of Δn_D are always positive for all the studied temperatures and for any composition.

The variation of $\Delta\sigma$ with composition of mixture (x_i of *p*-anisaldehyde) is presented in Figure 4. A detailed observation of the figure shows that the surface tension of a mixture

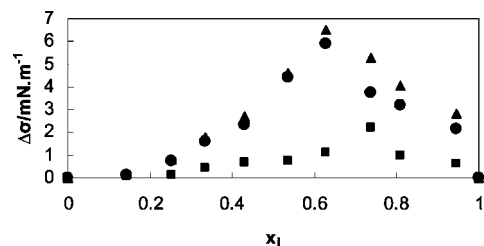


Figure 4. Deviations in surface tension for ($\Delta\sigma$) [*p*-anisaldehyde (1) + chlorobenzene (2)]: ■, $T = 303.15$ K; ●, $T = 313.15$ K; ▲, $T = 323.15$ K.

increases when mole fraction increases and shows an increasing trend when temperature increases.

Conclusion

Densities, viscosities, refractive indices, surface tensions, and ultrasonic velocities for a binary mixture (*p*-anisaldehyde + chlorobenzene) have been measured, and the corresponding excess molar volumes, viscosity deviations, refractive index deviation, and surface tension deviations were calculated at the temperatures of (303.15, 313.15, and 323.15) K. It has been observed that negative deviations were observed for viscosity deviations and positive deviations were observed for the deviations of refractive index, surface tension, and excess volume at (303.15, 313.15, and 323.15) K. It has been concluded that the Jouban–Acree model is very well suited for correlating the ultrasonic velocities and kinematic viscosity of the binary mixture studied. It is also clear that the Redlich–Kister polynomial equation can represent the excess molar volume, deviations of viscosity, refractive index, and surface tension very well which is indicated by low standard deviations.

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