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Experimental Study of the Thermophysical and Acoustical Properties of Acetophenone and Propylacetate

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Abstract Densities, viscosities, refractive indices and ultrasonic velocities of the binary mixtures of Acetophenone with Propyl acetate were measured over the entire mole fractions at (303.15, 313.15 and 323.15) K. From these experimental results, excess molar volumes V^E, viscosity deviation $\Delta\eta$, refractive index deviation Δn_D, deviations in isentropic compressibility ΔK_s and excess intermolecular free length ΔL_f are calculated. The viscosity values were fitted to the models of Krishnan- Laddha and McAllister. The thermo physical properties under study were fit to the Jouyban - Acree model. The excess values were correlated using Redlich-Kister polynomial equation to obtain their coefficients and standard deviations. It was found that in all cases, the data obtained fitted with the values correlated by the corresponding models very well. The results are interpreted in terms of molecular interactions occurring in the solution.

Keywords Viscosity, Density, Refractive Index, Ultrasonic Velocity, Molecular Interactions

1. Introduction

The thermodynamic, acoustic and transport properties of liquids and liquid mixtures[1] are used to study the molecular interactions between the various components of the mixtures and also to understand engineering applications concerning heat transfer, mass transfer, and fluid flow. 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 and ultrasonic velocity find extensive application in solution theory and molecular dynamics[2]. Such results are necessary for interpretation of data obtained from thermo chemical, electrochemical, biochemical and kinetic studies[3]. Acetophenone is an important industrial chemical widely used as an ingredient of flavour and fragrance in soaps, detergents, cosmetics and perfumes. Propyl acetate is used as a solvent in inks for coatings, cosmetics and personal-care products, intermediate for pharmaceuticals & agrochemicals, flexographic and rotogravure printing. Acetophenone and propyl acetate mixture was used in the manufacture of cosmetics amd fruit flavouring agents. In our earlier paper, we had studied the transport properties of binary liquid mixtures[4,5]. In continuation of this research, we have reported density (ρ), viscosity (η), refractive index (n_D) and sound speed (u) of pure acetophenone and propyl acetate 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 McAllister[6] and Krishnan and Laddha[7] model. The Jouyban –Acree[8] model has also been extended to density, viscosity, refractive index and sound speed (u) of binary mixtures. The deviation values have been fitted to Redlich-Kister[9] equation. Literature survey showed that no measurements have been previously reported for the mixture studied in this paper.

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2. Materials and Methods

All the chemicals used in this study were of analytical grade and obtained from Lobo Chemicals, India. The claimed mass fraction purity for the chemicals was ≥0.998. These chemicals were dried over molecular sieves and partially degassed prior to use[10, 11]. The purity of these experimental chemicals was checked by comparing the observed densities, viscosities, refractive indices and velocities with those reported in the literature. The measured values are included in Table 1 along with the available literature values.

Binary mixtures are prepared by mixing appropriate volumes of the liquid components in the specially designed glass bottles with air tight Teflon coated caps and mass measurements performed on a Shimadzu Corporation Japan type BL 2205 electronic balance, with a precision of ±0.01 mg. The required properties are measured on the same day immediately after preparing each composition. For all measurements, temperatures were controlled by circulating

the water through a thermostat (Technico,Madras. made in India) keeping temperature fluctuations within ±0.03K.

Table 1. Comparison of Experimental values of Pure Liquids with Literature Values at 303.15 K

Pure liquids	Acetophenone		Propyl Acetate	
	lit	Exp	lit	Exp
ρ / g ·cm-3	1.019412 1.016413	1.0199	0.876915 0.877714	0.8777
η/ mPa·s	1.45513	1.4553	0.518115 0.487814	0.4879
nD	1.522113	1.5221	1.379015	1.3792
u /ms-1	146012	1462	114916	1147

2.1. Density

Densities were determined by using a 25 cm^3 bicapillary pycnometer 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}.

2.2. Kinematic Viscosity

The kinematic viscosities were measured with Ostwald viscometer previously calibrated using water. The time was measured with a precision of 0.01s, 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

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

Where the two constants *a* and *b* were obtained by measuring the flow time *t* of benzene. The viscosities of mixtures of acetophenone and propyl acetate have been correlated with the model proposed by McAllister for a two-component mixture considering three body interactions.

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

In equation 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 masses of the components.

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

$$\ln\nu = x_1\ln\nu_1 + x_2\ln\nu_2 + x_1\ln M_1 + x_2\ln M_2 + \ln(x_1M_1 + x_2M_2 - 2.30x_1x_2(A_0 + A_1(x_1 - x_2)...)) \quad (3)$$

where A_0 and A_1 are interaction parameters. Jouyban et. al proposed a model for correlating the thermal properties of liquid mixtures at various temperatures

$$\ln y_{m,T} = f_1 \ln y_1 + f_2 \ln y_2 + f_1 f_2 \Sigma[A_J(f_1 - f_2)^J / T] \quad (4)$$

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.

2.3. Refractive Index

Refractive indices were measured using thermostatically controlled Abbe refractometer (Atago 3T) with accuracy less than 0.001units. Water was circulated in to the prism of the refractometer by a circulation pump connected to an external thermo stated water bath. Calibration was performed by measuring the refractive indices of doubly distilled water and propyl alcohol at defined temperatures. The sample mixture was directly injected in to the prism assembly of the instrument using a syringe. The solutions were pre thermo stated at the temperature of the experience before the experiments to achieve a quick thermal equilibrium.

2.4. Sound Speed

Speed of sound was measured by using a variable path, single crystal interferometer (Mittal Enterprises, New Delhi) at a frequency of 2MHz. 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 2 ms^{-1}. The isentropic compressibility was calculated by the equation

$$\kappa_s = 1/\rho u^2 \quad (5)$$

where ρ is the density of the mixture and u is the ultrasonic velocity of the mixture. The intermolecular free length (L_f) was calculated by the equation

$$L_f = K^* \kappa_s^{1/2} \quad (6)$$

where K= ((91.368+0.3565T) 10^{-8}) is temperature dependent Jacobson's constant.

3. Results and Discussion

Measured values of densities, viscosities, refractive indices and ultra sonic velocities of acetophenone with propyl acetate 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

$$V^E = (x_1M_1 + x_2M_2)/\rho m - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (7)$$

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

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (8)$$

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.

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

$$\Delta n_D = n_D - (x_1 n_{D1} + x_2 n_{D2}) \quad (9)$$

The isentropic compressibility deviation ($\Delta\kappa_S$) over the

entire composition range was obtained by

$$\Delta\kappa_S = \kappa_S - (x_1\kappa_{S1} + x_2\kappa_{S2}) \quad (10)$$

where x_1 and x_2 refer to the mole fraction of components 1 and 2. κ_{S1}, κ_{S2}, and κ_S refer to the isentropic compressibility of components 1 and 2 and the isentropic compressibility of the mixture, respectively.

Table 2. Densities ρ, Viscosities η, Refractive Indices n_D and Sound Speed u for the Acetophenone (1) + Propyl Acetate (2) Mixture at T= (303.15, 313.15, and 323.15) K

x_1	ρ/ g·cm^{-3}	η/ mPa·s	n_D	u /ms^{-1}
		303.15K		
0.0000	0.8777	0.4879	1.3792	1147.0
0.0896	0.8906	0.5836	1.3922	1175.5
0.1794	0.9035	0.6790	1.4052	1204.0
0.2695	0.9164	0.7744	1.4182	1232.3
0.3599	0.9294	0.8700	1.4312	1260.8
0.4506	0.9423	0.9656	1.4442	1289.2
0.5415	0.9552	1.0611	1.4571	1317.6
0.6326	0.9682	1.1567	1.4701	1346.0
0.7241	0.9811	1.2524	1.4831	1374.5
0.8158	0.9940	1.3482	1.4961	1403.0
0.9078	1.0070	1.4440	1.5091	1431.5
1.0000	1.0199	1.5140	1.5221	1460.0
		313.15K		
0.0000	0.8669	0.4327	1.3739	1123.0
0.0896	0.8799	0.5131	1.3869	1151.6
0.1794	0.8930	0.5935	1.3999	1180.2
0.2695	0.9060	0.6739	1.4130	1208.8
0.3599	0.9191	0.7543	1.4260	1237.4
0.4506	0.9322	0.8347	1.4390	1266.0
0.5415	0.9453	0.9151	1.4520	1294.6
0.6326	0.9584	0.9955	1.4650	1323.2
0.7241	0.9716	1.0759	1.4781	1351.8
0.8158	0.9847	1.1563	1.4911	1380.5
0.9078	0.9978	1.2367	1.5041	1409.2
1.0000	1.0110	1.3175	1.5172	1438.0
		323.15k		
0.0000	0.8558	0.3773	1.3686	1092.0
0.0896	0.8687	0.4597	1.3811	1122.3
0.1794	0.8816	0.5421	1.3935	1152.8
0.2695	0.8946	0.6245	1.4060	1183.3
0.3599	0.9076	0.7071	1.4186	1213.8
0.4506	0.9205	0.7896	1.4310	1244.4
0.5415	0.9335	0.8720	1.4435	1274.9
0.6326	0.9465	0.9545	1.4560	1305.3
0.7241	0.9596	1.0371	1.4685	1335.9
0.8158	0.9726	1.1196	1.4810	1366.5
0.9078	0.9856	1.2023	1.4935	1397.2
1.0000	0.9987	1.2855	1.5061	1428.0

The change of intermolecular free length (ΔL_f) on mixing were calculated by the equation

$$\Delta L_f = L_f - (x_1L_{f1} + x_2L_{f2}) \quad (11)$$

where L_{f1} and L_{f2} refer to the intermolecular free length of component 1 and 2.The excess molar volumes were fitted to a Redlich–Kister equation of the type

$$Y=x_1x_2 \Sigma A_i(x_1-x_2)^I \quad (12)$$

where Y is either V^E, and n is the degree of polynomial. Coefficients Ai were obtained by fitting equation 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

$$S(Y)=[\Sigma(A_{exp}-A_{cal})^2/(N-n)]½ \quad (13)$$

where N is the number of data points and n is the number of coefficients. The calculated values of coefficients along with the standard deviation (S) are given in Table 3.

Table 3. Parameters and Standard Deviations (S) of Redlich–Kister Equation for Acetophenone (1) +Propyl Acetate (2) T = (303.15, 313.15, and 323.15) K

Functions	A0	A1	A2	A3	S
		303.15 K			
V^E/cm^{3}mol^{-1}	0.0045	0.1194	-0.0014	-0.0798	0.0016
$\Delta\eta$/ mPa·s	0.0122	-0.0323	-0.0129	0.0202	0.0003
Δn_D	0.0004	-0.0049	-0.0004	0.0029	0.00004
$\Delta\kappa_s$ x10^{-11} m^{2}N^{-1}	-11.445	40.355	9.6089	-25.376	0.4418
ΔL_f X 10^{-11} m	1.052	-0.0931	-0.6565	0.1047	0.0088
		313.15K			
V^E/cm^{3}mol^{-1}	-0.0026	0.0578	0.016	-0.019	0.0008
$\Delta\eta$/ mPa·s	0.0107	-0.0253	-0.0116	0.0134	0.0002
Δn_D	0.0018	-0.0036	-0.002	0.0016	0.00003
$\Delta\kappa_s$ x10^{-11} m^{2}N^{-1}	43.908	-5.2928	-27.501	5.9452	0.4957
ΔL_f X 10^{-11} m	1.1288	-0.1013	0.6986	0.1142	0.0094
		323.15 K			
V^E/cm^{3}mol^{-1}	0.0107	0.015	-0.0018	0.0172	0.0003
$\Delta\eta$/ mPa·s	0.0119	-0.0183	-0.0121	0.0072	0.0001
Δn_D	0.0012	-0.0024	-0.0014	0.0006	0.00002
$\Delta\kappa_s$ x10^{-11} m^{2}N^{-1}	51.948	-6.6071	-32.418	7.3876	0.4435
ΔL_f X 10^{-11} m	1.3153	-0.1241	-0.8069	0.1387	0.0110

Table 4. Parameters and Standard Deviation of the of the McAllister model for Acetophenone + Propyl Acetate at T=(303.15, 313.15, 323.15)K

T/K	ν_{12}	ν_{21}	S
303.15	1.20410	0.96890	0.2854
313.15	1.03688	0.83261	0.2446
323.15	0.99166	0.80377	0.3475

Table 5. Parameters and Standard Deviation of the Krishnan – Laddha model for Acetophenone (1) + Propyl Acetate(2) at T =(303.15, 313.15, 323.15) K

T/K	A_0	A_1	A_2	A_3	S
303.15	-0.0071	0.000004	0.0169	-0.0114	0.3527
313.15	0.0097	-0.0007	-0.0077	-0.0045	0.2905
323.15	-0.3142	-0.0046	0.04463	-0.0076	0.3192

Table 6. Parameters and Standard Deviations of Jouyban- Acree Model for Acetophenone (1) + Propyl Acetate (2)

Properties	T/K	A0	A1	A2	A3	S
ρ/ g·cm^{-3}	303.15	4.2199	-0.1202	-3.9253	-0.5297	0.00008
	313.15	4.3539	-0.0899	-3.9851	-0.6843	0.00007
	323.15	4.4689	-0.1035	-4.1089	-0.6667	0.00007
η/ mPa·s	303.15	210.17	-37.32	-189.27	-235.46	0.00584
	313.15	196.05	-36.049	-180.99	-219.09	0.00463
	323.15	241.9	-47.258	-226.54	-302.29	0.00505
n_D	303.15	2.0038	-0.0357	-1.7894	-0.286	0.00005
	313.15	2.0633	-0.0032	-1.8595	-0.6326	0.00006
	323.15	1.9739	-0.0052	-1.693	-0.47	0.00005
u m/s	303.15	10.153	-0.7027	-8.9832	-2.5673	0.26331
	313.15	10.89	-0.6776	-9.9063	-3.0658	0.26949
	323.15	12.93	-1.0455	-11.772	-2.3135	0.27447

Interaction parameters and standard deviations of the McAllister model and Krishnan and Laddha model for the viscosity of acetophenone and propyl acetate mixture at (303.15, 313.15, and 323.15) K are presented in Table 4 and Table 5. Constants and standard deviations of the Jouban-Acree model of the acetophenone and propyl acetate at (303.15, 313.15, and 323.15) K are presented in Table 6.

The variation of excess volumes with the mole fraction (x_1) of acetophenone and propyl acetate at (303.15, 313.15 and 323.15) K are represented in figure.1. This shows that the excess molar volumes are always negative for all the studied temperatures. Treszczanowicz et al.[17] and Roux and Desnoyers[18] suggested that V^E is the resultant contribution from several opposing effects. These may be divided arbitrarily into three types, namely chemical, physical and structural. A physical contribution, that is specific interactions between the real species present in the mixture, contribute a negative term to V^E. The chemical or specific intermolecular interactions result in a volume decrease, and these include charge transfer type forces and other complex forming interactions. This effect contributes negative values to V^E. The structural contributions are mostly negative and arise from several effects, especially from interstitial accommodation and changes of free volume. In other words, structural contributions arising from geometrical fitting of one component into the other due to the differences in the free volume and molar volume between components lead to a negative contribution to V^E.

The variation of viscosity deviations, with the mole fraction of component 1 is presented in figure 2. Viscosity values are positive for the acetophenone and propyl acetate mixture at all the studied temperatures. Figure 2 shows that the viscosity deviations are positive[19], indicates that the interaction between binary mixtures is strong.

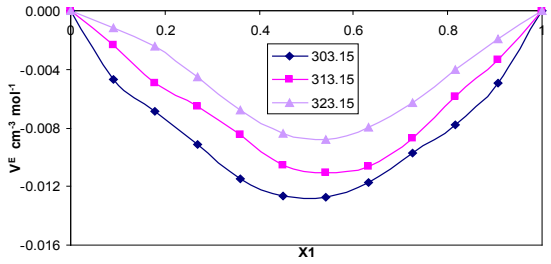

Figure 1. Excess Molar Volume, V^E, for the system Acetophenone (1) + Propyl Acetate (2) at temperatures: ♦, T= 303.15 K; ■, T= 313.15 K; ▲, T= 323.15 K

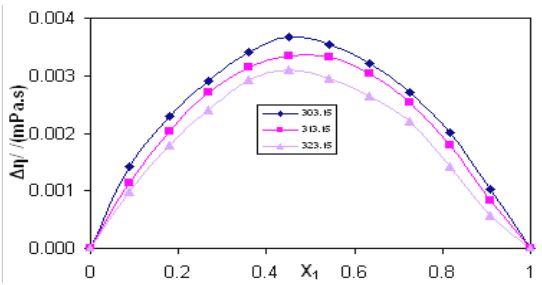

Figure 2. Viscosity Deviation, Δη, for the system Acetophenone (1) + Propyl Acetate (2) at temperatures: ♦, T= 303.15 K; ■, T= 313.15 K; ▲, T= 323.15 K

The results of refractive indices versus x_1 at (303.15, 313.15 and 323.15) K for the systems of acetophenone are shown in figure 3. Here the system acetophenone + propyl acetate exhibit a positive deviation at all the studied temperatures. The values of $\Delta\kappa_s$ are negative at all the temperatures and the values of $\Delta\kappa_s$ become less negative as temperature increased. This may be attributed to the weakening of structure making interactions at elevated temperatures due to enhanced thermal motion. The excess free length is negative over the whole mole fraction range for all binary mixtures at different temperatures, figure 5. This indicates structural readjustment in the liquid mixtures towards less compressible phase of fluid and closer packing of molecules[14,20].

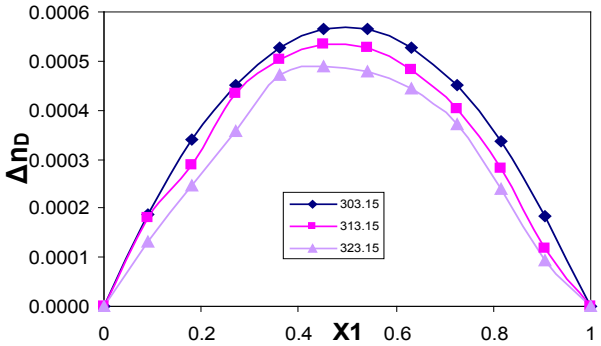

Figure 3. Refractive Index Deviation, Δn_D , for the system Acetophenone (1) + Propyl Acetate (2) at temperatures: ♦, T= 303.15 K; ■,T= 313.15 K; ▲,T= 323.15 K

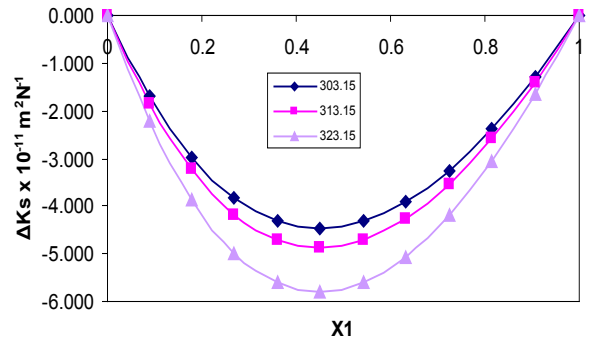

Figure 4. Isentropic Compressibility Deviation, $\Delta\kappa_s$, for the system Acetophenone (1) + Propyl Acetate (2) at temperatures: ♦, T= 303.15 K; ■,T= 313.15 K; ▲,T= 323.15 K

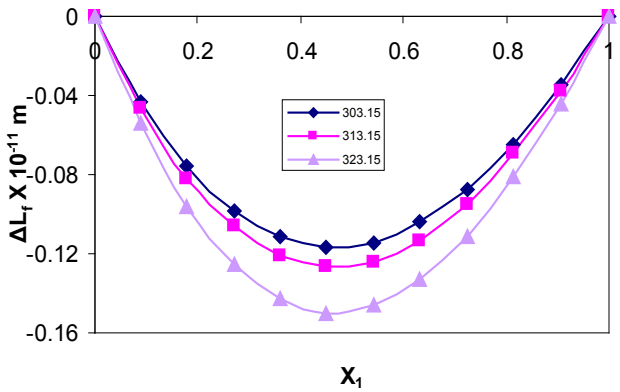

Figure 5. Intermolecular Free Length Deviation, ΔL_f , for the system Acetophenone (1) + Propyl Acetate (2) at temperatures: ♦, T= 303.15 K; ■,T= 313.15 K; ▲,T= 323.15 K

4. Conclusions

Densities, viscosities, refractive indices and ultrasonic velocities for a four binary mixtures have been measured. Excess molar volumes, viscosity deviations, refractive index deviations, compressibility deviation and change in intermolecular free length for mixtures of acetophenone and propyl acetate were obtained from the experimental results and fitted by the Redlich Kister equations. It has been concluded that the Jouyban Acree model is very well suited for correlating the thermo physical properties of the binary mixture studied.

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