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Address:-Ashok Yakkaldevi 258/34, Raviwar Peth, Solapur - 413 005 Maharashtra, India
Cell : 9595 359 435, Ph No: 02172372010 Email: ayisrj@yahoo.in Website: www.aygrt.isrj.org

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ULTRASONIC STUDIES ON MOLECULAR INTERACTIONS IN BINARY AND TERNARY MIXTURES OF BENZOPHENONE WITH n-ALKANOLS AND TOLUENE

G.R.Balaji[1] , M.Prabhakaran[1] , S.Rajasekar[1] and P.S.Joseph[2]

[1]Department of Physics, Syed Ammal Engineering College, Ramanathapuram , India.
[2]Department of Physics, Cauvery College of Engineering & Technology, Tiruchirappalli, India .

Abstract:- The ultrasonic velocity (u), density (ρ) and viscosity (η) at 302 K have been measured in the binary systems and ternary systems of benzophenone in 2-propanol, 1-butanol and toluene. The acoustical parameters such as adiabatic compressibility (βad), bulk modulus (k), acoustic impedance (Z), intermolecular free length (L_f), relaxation time (t), relative association (RA), Gibb's free energy (? G^{*E}) and attenuation (a/f^2) were calculated and different types of molecular interactions in these mixtures have been analyzed on the basis of the variation of those parameters with increasing concentration of benzophenone and keeping constant ultrasound frequency at 2MHz.

Keywords: Ultrasonic velocity, Density, Viscosity, Acoustic parameters, benzophenone,2-propanol, 1-butanol, toluene.

1. INTRODUCTION

During the past decade, an increasing interest in benzophenone and its derivatives emerged, accompanied with research activities in various fields. The reason for this is that of benzophenones are used as a photo initiator of UV-curing applications in inks, adhesive and coatings, optical fiber as well as in printed circuit boards. Some benzophenone derivatives are excellent optical filters and, therefore, current industrial applications involve their use as sunscreen agents for the production of skin-protectors and their addition to plastic casings, which are used to prevent damaging of scents and colors by ultraviolet radiation in products such as perfumes and soaps[1].Benzophenone and its derivatives are also used as NLO material[2].In chemical industry, the knowledge of the thermodynamic properties of solutions is essential in the design involving chemical separation, heat transfer, mass transfer and fluid flow[3].

The ultrasonic velocity of a liquid is fundamentally related to the binding forces between atoms or molecules, and has been successfully employed in understanding the nature of molecular interactions in pure liquids and binaryandternarymixtures [4-6] and such studies assist us to detect and assess weak and strong molecular interactions, present in binary and[7-8]ternary[9] liquid mixtures.Many researchers have reported that the acoustical properties of synthesized compounds in various solvents and also thermodynamic properties of binary mixtures of 1, 3, 4–Oxidazolederivative[10-12].The applications of benzophenone derivatives attract us to study their behavior in various solvent and also their thermodynamic properties with the help of ultrasound. In this paper, the thermodynamic properties of the title compound benzophenonewithn-alkanols and toluenewereinvestigated and reported.

2. EXPERIMENTAL

2.1 Materials and sample preparation

The commercially available AR grade chemicals were used for the present study and quoted mass fraction

purities of the materials were:Benzophenone (>99%), 2-propanol (>99.5%), 1-butanol (>99.5%) and toluene (>99.7%) and were purified by standardprocedure.The purity of the chemicals were verified by measuring the densities (?), sound velocity (u)and viscosity (?) which were in good agreement with literature values[13-14]. In all systems, the mixtureswere prepared by changing the concentration (molarity) of benzophenone from 0.1M to 0.6M in the step of 0.1M with fixed volume of solvents (say 40ml) so as to have the binary and ternary mixtures of benzophenone+2-propanol, benzophenone+1-butanol, bezophenone+toluene+2-propanol and bezophenone+ toluene+1-butanol for the present investigation. The preparedsolutions are stored in air tight stoppered glass bottle to avoid any contamination and evaporation and all the solutions were leftfor 1 hour and completesolubility is found.

2.2 Methods

2.2.1 Density measurements

The density measurements were performed using a specific gravity bottle made of borosil glass having single capillary and a bulb of capacity 25ml. The density of the pure liquids and liquid mixtures were calculated using relative measurement method. A clean dry specific gravity bottle was weighed (w_1) using anelectronic balance (SHIMADZU-ELB 300) with an accuracy of ±0.1 mg. The specific gravity bottle was filled with air bubble free deionizeddouble distilled water and kept in constant temperature bath to attain the experimental temperature of 302K and it is taken as reference liquid. The weight of the specific gravity bottle with water (w_2) was measured. Thus the weight of the water was determined as $w_w = w_2 - w_1$. Thedensities of liquid mixtures were found out by using the relation [15] ?= (w/w_w) ?$_w$.Where 'w' is the weight of the liquid mixture, 'ρ_w' is the density of water. The accuracy in the density measurement was ±0.10 kgm^{-3}.

2.2.2 Viscosity measurements

The viscosity of pure liquids and liquid mixtures were determined by relative method, using 10 mlOstwald's viscometer. Initially the viscometer was filled with deionizeddouble distilled water at room temperature. The time flowof water (t_w) from first mark to second mark was noted. The same procedure was repeated for the liquid mixtures also and the time flow of liquid mixture (ts) was measured and the viscosity of liquid mixtures were determined using the following relation[16]?=?$_w$(?$_s$t$_s$/?$_w$t$_w$)

Here,'η_w', 'ρ_w' and 't_w' are viscosity, density and time flow for the double distilled water respectively. '?$_s$' and 't_s'are density and time flow of the liquid systems. The accuracy of the measurement of viscosity is ±0.001 Nsm^{-2}.The measurements were repeated five times and the average values are reported.

2.2.3 Ultrasonic measurements

The ultrasonic velocity (u) in pure liquids and liquid mixtures have been measured using an ultrasonic interferometer (F81, Mittal Enterprises, New Delhi, India) operated at single frequency 2MHz with an overall accuracy of ±0.1ms^{-1}. The temperature of the doublejacketed interferometer cell was maintained by circulating water from a constant temperaturebath. All the precautions were taken to avoid the possible experimental error and human error.The following acoustical parameters were calculated by using the experimental data.

Ultrasonic velocity [17] :u=f?
Where, 'f' is the frequency of ultrasonic wave.
Adiabatic compressibility [18]:ß$_{ad}$=(?u^2)$^{-1}$
Where, 'ρ' is density of the solution and 'u' is ultrasonic velocity in solution.
Bulk modulus [19]: k=?u^2
Acoustic Impedance [20]: Z=u?
Intermolecular free length [21]: $L_f = k_f/(?u)^{1/2}$
Temperature dependent Jacobson's constant k_f= (93.875+0.375T) x 10^{-8}. Where 'T' is aabsolute temperature.
Relaxation time [22]: t = 4?/(3?u^2)
Relative association [23]: $R_A = (?/?_0)(u_0/u)^{1/3}$
Where, 'ρ','ρ_o','u_o', and 'u' are the density of solution, density of the solvent, ultrasonic velocity in solvent and ultrasonic velocity in solution respectively.
Gibb's free energy [24]:? G=(k_BT)log(k_BTt /h)
Where,'K_B'is the Boltzmann's constant (1.3806×$10^{-23}$$JK^{-1}$), 'h' is the Plank's constant
(6.63 x 10^{-34}JS), 'T'is the absolute temperature and 't' is the relaxation time.
Attenuation [25]: (af^2)=8p^2?/(3?u^3)

Where,'a' is the absorption co-efficient,'η' is viscosity.

3. RESULT AND DISCUSSION

The experimentally observed values of ultrasonic velocity (u), density (ρ) and viscosity (?) and calculated acoustical parameters such as adiabatic compressibility ($ß_{ad}$), bulk modulus (k), acoustic impedance (Z), intermolecular free length (L_f), relaxation time (t), relative association (RA), Gibb's free energy (? G*E) and attenuation (a/f^2) are listed in Tables 1 and 2.

From Table1, it is found that the density and viscosity of all liquid mixtures were found to be increased with increasing concentration of benzophenone. Due to the increase of benzophenone molecules in solution,the medium became denser[26]. Further, theincrease in the number of particles in solution is responsible for increasing the cohesive force between the liquid layers.Thereby, the co-efficient of viscosity increases in all systems.

The Figure 2 reveals that the ultrasonic velocity increases in all four systems with increasing concentration of benzophenone. This may be due to the association of very strong dipole-dipole interaction between the constituent molecules. Hence this behavior can be attributed to study the intermolecular interaction in the system[27] and proposed that the ultrasonic velocity depends upon the increase or decrease of inter molecular free length. The ultrasonic velocity should be increased by decreasing the intermolecular free length and vice-versa [28] and it has been observed in the present investigation in all liquid mixtures.

As ultrasonic velocities increases in all systems with the concentration of benzophenone, the adiabaticcompressibility ($ß_{ad}$) and intermolecular free length (L_f) show the reverse trend. From the Figures 3 and 4, the both parameters are found to be decreased in all four systems with different concentration of benzophenone.The decrease in adiabatic compressibility and intermolecular free length with increasing the concentration of benzophenone pointed out the considerable molecular interaction between benzophenone and alkanol molecules forming hydrogen bonding through dipole-dipole interaction. The decrease in adiabatic compressibility with increasing concentration indicates that the presence of intermolecular forcesand which brings the molecules to a closely packing and resulting into decreasing in intermolecular free length.Since ultrasonic is a mechanical wave, it can create compressive force per unit area in liquid during the propagation.

From Table 2, it is found that the bulk modulus increases against different concentration persist in all systems and confirmed that the decreasing adiabatic compressibility and intermolecular free length.Alkanols are the liquids and as they are associated through a three dimensional network of hydrogen bond[29] and can be associated with any of other group having some degree of polar attraction[30].In binary mixture, when alkanols are added with benzophenone, the hydroxyl group (O-H) of alcohols and the carbonyl groups (C=O) of aromatic compound may interact with each other and forming hydrogen bonding through dipole-dipole interaction. The aromatic groups can create possible interaction between the p electrons cloud and hydroxyl group. This interaction is rather weak than hydrogen bonding, though it can lead to form intermolecular complexes [31]thereby, the ultrasonic velocity increases with increasing concentration of benzophenone.In the case of ternary mixtures, the observed ultrasonic velocity were found to be larger than binary mixtures against different concentration persists. Therefore, very strong molecular interaction has been identified by adding of toluene with alcohols and bezophenone.The p electrons from the toluene ring may be withdrawn by carboxyl group of ketone and which acts as electron-acceptor towards p electrons of toluene ring [32].This observation indicates that the presence of multitype of interaction among different species involved in the system like dipole-dipole interaction through hydrogen bonding formation between benzophenone and alkanols and donor-acceptor character between toluene and benzophenone.

The inertial and elastic properties of the medium can be related by using the acoustic impedance parameter and it is important to studytheacoustic impedance in the relation to theconcentration[33]. Table 2 reveals that the variation of acoustic impedance with concentration. The acoustic impedance shows an increasing trend with concentration similar to the manners of ultrasonic velocity. Such an increasing trend of acoustic impedance making more reliableto understand the molecular interactions of dipole-dipole interaction through hydrogen bonding formation between solute and solvent [34].

The variation of relaxation time with concentration is shown in Figure 5. It brings out that the relaxation time decreases non-linearly with concentration of benzophenone in binary mixtures and increases non-linearly in ternary mixtures. Owing to having keen look in table, the ultrasonic relaxation time of binary mixtures was found to be larger than ternary mixtures. This clearly indicates that, the pressure of the molecular interaction in the system can be studied and it may be due to the specific interaction between the unlike molecules present in the system and intermolecular force with the addition of solute in solvent[35].

The Figure 6 shows that the variation of relative association (R_A) with concentration. If R_A decreases with increasing concentration which can indicate the breaking up solute molecules in solution [36] but in our present investigation, the relative association increases with increasing concentration in all systems. It may be due to the H-bond formation in liquid mixtures. Herethe relative associations were found to be larger in the case of ternary mixture than binary mixture which shows that the strong solute solvent interactions exist in ternary mixtures than binary

mixtures.

The Figure 7shows thatthe variation of Gibb's free energy with concentration. The temperature and relaxation time contribute in this Gibb's free energy computation. In this present study, the Gibb's free energy decreases nonlinearly in binary mixtures and increases nonlinearly in ternary mixtures. The lessening of Gibb's free energy in binary system shows that the requirement for tiny time for making the rearrangement of molecules in the mixture and decreasing the energy that leads to dissociation [37]. In the case of ternary mixture, the Gibb's free energy increases nonlinearly due to the association of molecules. The attenuation coefficient also decreases nonlinearly in binary mixtures and increases nonlinearly in ternary mixtures.The viscosity contributes a major role in the measurements of ultrasonic attenuation. The nonlinear variations may be due to the molecular association and dissociation. The nonlinearity of these plots indicates their dependence on molecular structure and interactions occuramong the molecules of the element of mixtures [38-39].

4. CONCLUSION

The experimental observation of ultrasonic velocity, density and viscosity and their variation indicates the presence of molecular interaction in the title compounds. The direct parameters like adiabatic compressibility, acoustic impedance, inter molecular free length, ultrasonic relaxation time, relativeassociation,Gibb's free energy and attenuation were also confirmed that the existence of molecular interaction in the systems. It has been identified that the molecular interactions in the ternary mixtures were stronger than that of the binary mixtures and a competitive mechanism established in H-bond formation and dipole-dipole interaction between the solute and solvent used in this investigation.

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Table 1: Experimental values of ultrasonic speeds (u), density (ρ) and viscosity (η) at 302K for system 1, 2, 3 and 4.

X	benzophenone+2-propanol			benzophenone+1-butanol			Benzophenone +toluene+ 2 propanol			Benzophenone +toluene+1-butanol		
mol/lit	Velocity *u* (m/s)	Density r (kg/m^3)	Viscosity $hx10^{-3}$ (Nsm^{-2})	Velocity *u* (m/s)	Density r (kg/m^3)	Viscosity $hx10^{-3}$ (Nsm^{-2})	Velocity *u* (m/s)	Density r (kg/m^3)	Viscosity $hx10^{-3}$ (Nsm^{-2})	Velocity *u* (m/s)	Density r (kg/m^3)	Viscosity $hx10^{-3}$ (Nsm^{-2})
0.1	1130.60	781.76	1.485	1231.00	808.37	1.890	1196.66	821.84	0.7183	1241.33	834.97	0.8354
0.2	1140.00	786.73	1.487	1235.20	813.69	1.903	1201.80	824.68	0.7480	1246.85	839.94	0.8421
0.3	1145.60	791.69	1.499	1239.11	818.30	1.933	1203.00	831.42	0.7572	1252.00	844.90	0.8569
0.4	1154.66	795.95	1.538	1242.00	823.62	1.940	1216.00	837.10	0.7946	1256.00	848.09	0.8823
0.5	1160.00	802.69	1.551	1246.66	828.59	1.976	1224.00	842.77	0.8121	1265.00	852.70	0.9150
0.6	1172.92	810.14	1.589	1248.00	831.78	1.990	1230.80	844.19	0.8175	1268.00	855.54	0.9397

Table 2 The values of adiabatic compressibility (β_{ab}), Bulk modulus (k),Acoustic impedance (Z), Intermolecular free length (L_f), Relaxation time (τ). Relative association (R_A), Gibb's free energy (ΔG^{*E}) and attenuation(a/f^2)at 302K for the systems 1, 2, 3 and 4.

Concentration (X) mol/lit	Adiabatic compressibility (β_{ad}) ($X10^{-10}$ m^2N^{-1})	Bulk modulus (k) ($x10^9$ Nm^{-2})	Acoustic impedance (Z) ($x10^5Kgm^{-2}s^{-1}$)	Intermolecular free length (L_f) ($X10^{-09}$ m)	Relaxation time (τ) ($X10^{-12}$ S)	Relative association (R_A)	Gibb's free energy (ΔG^{*E}) ($x10^{-20}$kJ mol^{-1})	Attenuation (α/f^2) ($X10^{-14}Npm^{-1}s^{-2}$)
System 1: benzophenone+2-propanol								
0.1	10.0071	0.9993	8.8386	2.20712	1.98127	1.005031381	1.0517	3.45561
0.2	9.78058	1.0224	8.9687	2.19105	1.93669	1.008633213	1.0422	3.34999
0.3	9.62451	1.0390	9.0696	2.17883	1.92298	1.013335666	1.0393	3.31002
0.4	9.42336	1.0612	9.1905	2.16445	1.93192	1.016116688	1.0412	3.29931
0.5	9.25840	1.0801	9.3112	2.15038	1.91439	1.023146195	1.0374	3.25433
0.6	8.97229	1.1145	9.5023	2.12865	1.90105	1.028836703	1.0345	3.19605
System 2: benzophenone+1-butanol								
0.1	8.16345	1.2250	9.9510	2.08010	2.05719	1.010182888	1.0674	3.29538
0.2	8.05502	1.2415	10.0507	2.06976	2.04383	1.015677243	1.0647	3.26284
0.3	7.95917	1.2564	10.1396	2.06066	2.05107	1.020356107	1.0662	3.26407
0.4	7.87100	1.2705	10.2294	2.05161	2.03544	1.026192547	1.0630	3.23166
0.5	7.76541	1.2878	10.3297	2.04162	2.04593	1.031096985	1.0651	3.23617
0.6	7.71902	1.2955	10.3806	2.03660	2.04811	1.034696030	1.0656	3.23615
System 3: benzophenone+toluene+2-propanol								
0.1	8.49711	1.1769	9.8346	2.09237	0.813796	1.036747160	0.68083	1.34102
0.2	8.39557	1.1911	9.9110	2.08430	0.837318	1.038844552	0.69271	1.37388
0.3	8.31090	1.2032	10.0020	2.07479	0.839069	1.046986533	0.69358	1.37538
0.4	8.07896	1.2378	10.1791	2.05666	0.855939	1.050369223	0.70188	1.38803
0.5	7.92005	1.2626	10.3155	2.04302	0.857583	1.055174853	0.70268	1.38161
0.6	7.81961	1.2788	10.3903	2.03566	0.852337	1.055002636	0.70012	1.36557
System 4: benzophenone+toluene+1-butanol								
0.1	7.89657	1.2664	10.2017	2.05438	0.756280	1.024158944	0.70662	1.20139
0.2	7.79985	1.2821	10.2825	2.04630	0.777905	1.026179253	0.70378	1.23027
0.3	7.67310	1.3033	10.4094	2.03379	0.774676	1.033145595	0.70515	1.22013
0.4	7.57257	1.3206	10.5140	2.02365	0.802289	1.039098297	0.71310	1.25959
0.5	7.41498	1.3486	10.6610	2.00964	0.802894	1.043649646	0.72005	1.25158
0.6	7.36752	1.3573	10.7043	2.00557	0.803060	1.044583006	0.72780	1.24887

Figure 1.plots of Concentration(x) versus Density (ρ) of binary and ternary mixture at 302K

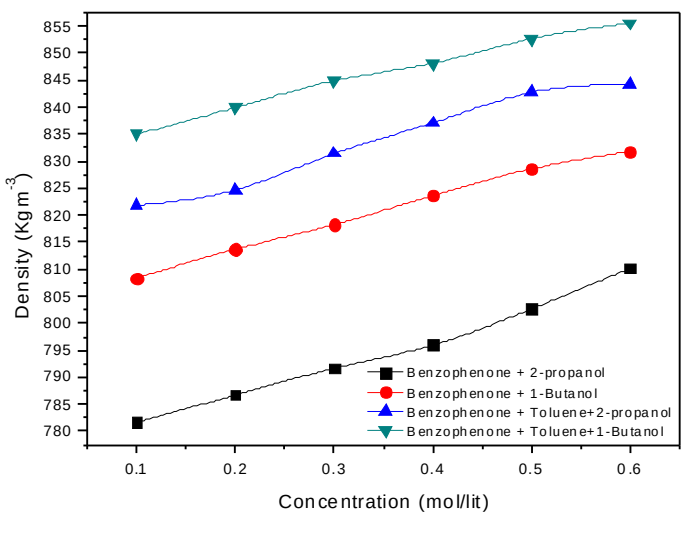


Figure 2.Plots of Concentration(x) versusUltrasonic velocity (u) of binary and ternary mixture at 302K

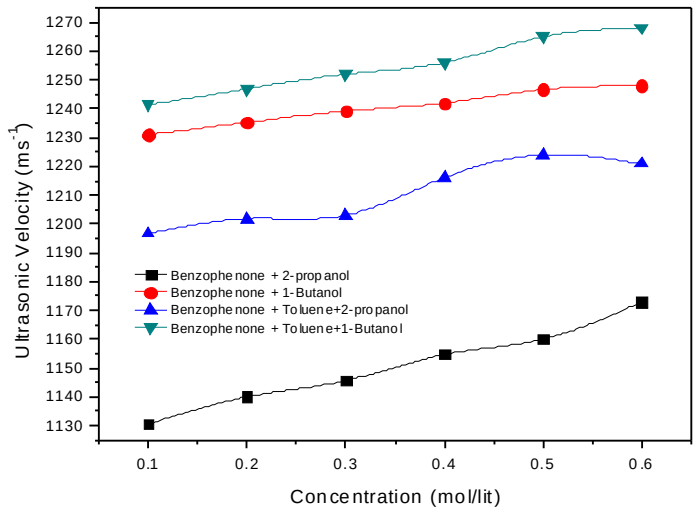


Figure 3. Plots of Concentration (x) versusAdiabatic compressibility (β_{ad}) of binary and ternary mixture at 302K

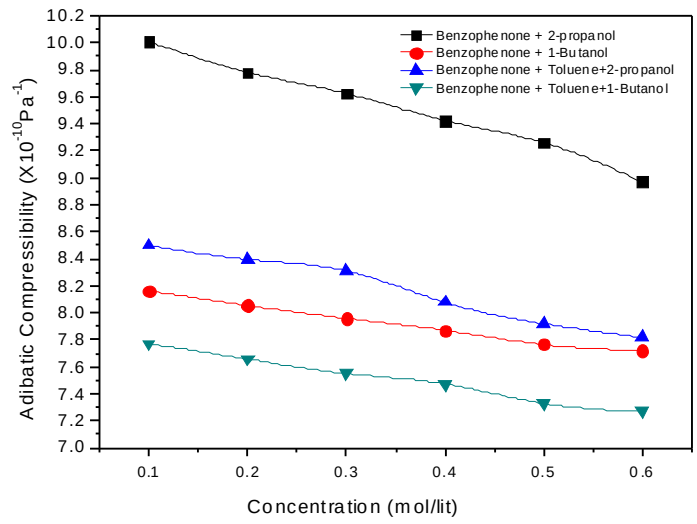


Figure 4.Plots of Concentration (x) versus free path length (L_f) of binary and ternary mixture at 302K

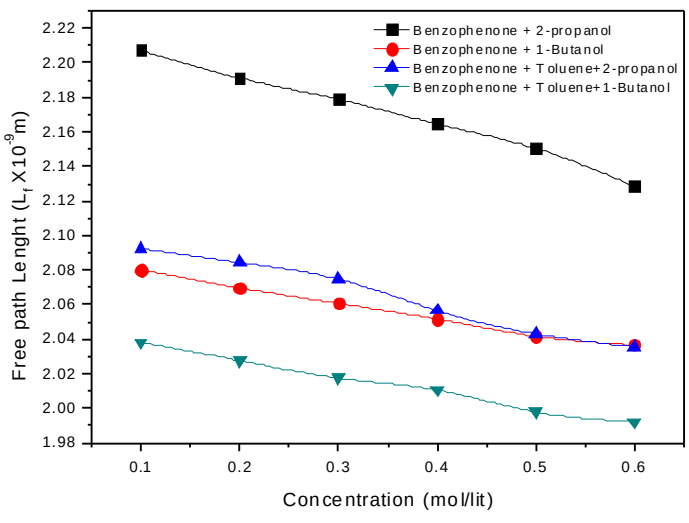


Figure 5.Plots of Concentration (x) Relaxation time (τ) of binary and ternary mixture at 302K

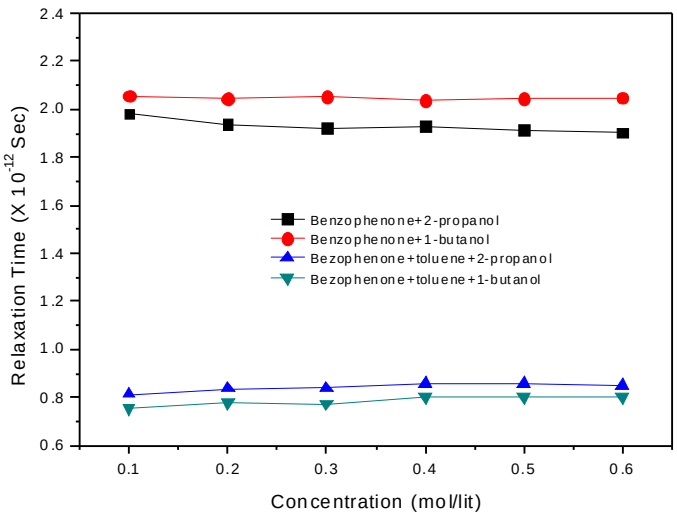


Figure 6.Plots of Concentration (x) versusRelative association (R_A) of binary and ternary mixture at 302K

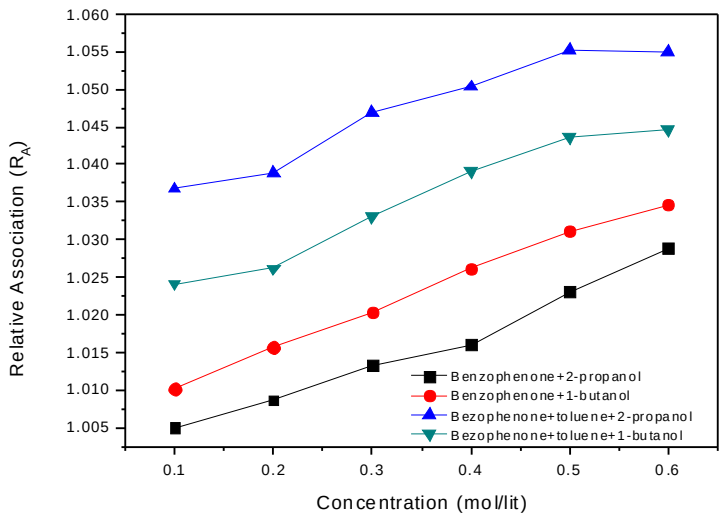
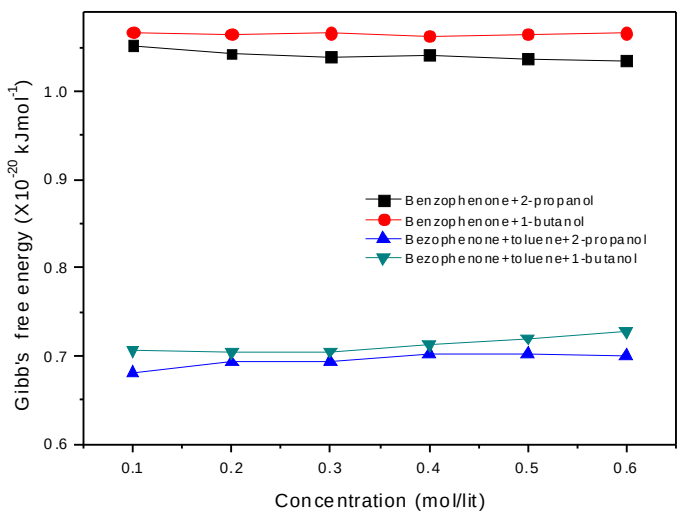


Figure 7.Plots of Concentration (x) versusGibb's free energy (ΔG^{*E}) of binary and ternary mixture at 302K



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G.R.Balaji
Department of Physics, Syed Ammal Engineering College, Ramanathapuram , India.