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DIELECTRIC BEHAVIOUR OF ALLYL AMINE (AA) AND 2-METHOXY ETHANOL (2-ME), AT 9.85 GHZ MICROWAVE FREQUENCY

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Abstract
The liquid dielectrics mainly serve as impregnates for high voltage cables, capacitors and act as heat transfer agents that is for cooling in transformers.Using Surber's technique of measuring reflection coefficient from air dielectric boundary of the liquid dielectric constant (έ) and dielectric loss (ε̋) of Allyl Amine (AA) with 2-Methoxy Ethanol for different mole fractions of AA, have been measured at single microwave frequency 9.85 GHz at 30^0C. Density (ρ), viscosity (η) and squared refractive index (n^2_D), including pure liquids are reported. Dielectric constant (έ) and Dielectric Loss (ε̋) used to evaluate loss tangent (tanδ), molar polarization (P_{12}) and excess dielectric parameters. Dielectric and excess parameters are being used to explain the formation of complexes in the system. The intermolecular hydrogen bonding interaction between Allyl Amine (AA) AND 2-Methoxy Ethanol (2-ME), has been investigated by FT-IR spectroscopy.The information about dielectric behaviour of material gives superior understanding in the selection of solid and liquid insulating materials. These results confirm that the mixtures form hydrogen-bonded structures, which are strongly influenced by the numbers of hydroxyl groups and carbon atoms of the alcohol molecules and vary with the concentrations of the mixtures.

Keywords: Complex formation, X-band microwave bench, Allyl Amine (AA), 2-Methoxy Ethanol (2-ME)

Introduction
Dielectric analysis of binary mixtures exhibited composition and formation of complexes by interacting between the molecules. Molecules change its

characteristics when it forms the complexes. Dielectric evaluations assisted for detection of the interaction between molecules along with its orientation and thermodynamic properties[1]. According to literature, researcher paid attention towards the amines as one of the constituent components in the binary mixtures and suggest the strong interaction between the alcohols and amine molecules.[2-8] Bhupesh G. Nemmaniwar[5] observed solute-solvent molecular associations in 2-chloroaniline (2CA) + 2-methoxyethanol (2ME) and 2-chloroaniline (2CA) + 2-ethoxyethanol. U.Tumberphale[9] investigated the significant role of dielectric of binary mixtures for the multiple complex formations formation and they also proposed the strong interaction between 2-Alkoxyethanols with Aniline molecules. The formation of 1:1 complexes in ethyl methyl ketone and ethylenediamine and methyl isobutyul ketone and ethylenediamine at 9.44 GHz frequencies at 30^0C[4]. The heterogeneous interaction between the unlike molecules when studied the complex permittivity spectra of 2-butoxyethanol in anilines[10]. When thermodynamic evaluation of the binary mixture of diisopropylamine (DIIPA) + 2-methyl-1-propanol+2-propanol and + 1-butanol evaluated, the presence of strong molecular interactions was observed between the molecules[11]. In both systems N, N-dimethylacetamide + methanol N,N-dimethylacetamide + ethanol [7] . Multiple frequency capacitance-voltage (C-V) measurements of Allyl amine (AA) with water was carried out by Yifan Xu et al 2004[11]. They observed that during film deposition, the insulator dielectric constant was increased by radio frequency (RF) pulsed plasma polymerization[11]. Dielectric Constant measurements are not possible with deposited film layers, which are very thin insulating sheets sometimes used to prevent material from adhering to a mold or platen[12].

With this background, present paper is focused on the molecular interaction between binary mixtures of polar liquids. The possible formation of AA and 2-ME complex may be due to molecular association between these liquids. The aliphatic 2-ME is amphoteric in nature having (-O-) and hydroxyl (-OH-) group in the same molecule[13] while Allyl amine is a stable organic compound and the simplest unsaturated amine having extensive pharmaceutical applications. The presence of (-O-) and (-OH-) groups in 2-ME form intra and intermolecular hydrogen bonds disturb the binary mixture condition Because ,in solid states, the amino group of aniline's derivatives has more than two absorption bands.[14] In physiologically important systems, hydrogen bonding is responsible for biological activity. Hydrogen bonding is formed between functional group and an atom or group of atoms in the same or different molecules[15-17]. Hence, FTIR spectroscopic studies have been carried out on the binary mixture of AA and 2-ME.Therefore, the dielectric behaviour of 2-ME and AA mixture is given deep sense of interest to analyse the molecular interactions and the formation of complexes in the mixture.

Materials and Methods:

AR grade Allyl Amine (AA) and 2-Methoxy Ethanol (ME) from S.D fine chemicals where used without further purification.

Binary mixtures with different mole fraction of Allyl Amine in the mixture were prepared and kept for six hours in well stoppered bottles to ensure good thermal equilibrium. The density (ρ) and viscosity (η) of pure components and their mixtures were measured at room temperature by pykometer and oswals's vicometer, respectively.

The refractive indices for Sodium-D-lines were measured by Abbe's Refractometer. The Dielectric constant measurements were carried out from the X-band microwave bench of oscillating frequency 9.85 GHz using Surber's technique at 30^0C[18]. FTIR spectra of polar mixtures in different concentration were carried out by Thermofischer Scientific Nicolet iS5.

Calculations:

The values of (έ) and (ε̋) for low loss liquids are calculated according to Hestone, W. H.et al ;1950[19]

$$\acute{\varepsilon} = [\frac{\lambda_0}{\lambda_c}]^2 + [\frac{\lambda_0}{\lambda_d}]^2 \quad \text{----------(1)}$$

$$\ddot{\varepsilon} = \frac{2}{\pi}[\frac{\lambda_g}{\lambda_d}][\frac{\lambda_0}{\lambda_d}]^2[\frac{\partial\rho}{\partial n}] \quad \text{----------} \quad (2)$$

The free energy of activation E_a is obtained using relation[20]

$$\eta = \frac{hN}{V} e^{\frac{Ea}{RT}} \quad \text{----------} \quad (3)$$

Where,

η is viscosity and V is molar volume of the liquids.

And Molar polarization were obtained using[21]

$$P_{12} = (\frac{\acute{\varepsilon}-1}{\acute{\varepsilon}+2})[\frac{M1x1+M2x2}{\rho}] \quad \text{----------} \quad (4)$$

The excess parameters are calculated by the formula defined by

$$\Delta Y = Y_m - (X_1Y_1 + X_2Y_2) \quad \text{----------} \quad (5)$$

Where,

ΔY-Excess parameter and Ym Dielectric parameters for mixture.

The subscripts m, 1 and 2 represents mixture, component 1 and component 2 respectively.

X_1 and X_2 is mole fraction of the 2 components.

The values of Mole fraction (x) of Allyl Amine AA and (X_B) 2-ME, density (ρ), refractive index (n), dielectric constant (έ), dielectric loss (ε̋), loss tangent (tan δ), activation Energy (E_a) and molar polarization (P_{12})for binary mixture measured at 30^0 C which is represented in Table-1.

The excess dielectric parameters provide significant information regarding interaction between the polar-polar liquid mixtures. The values of Mole fraction (x) of Allyl Amine AA and 2-ME and its binary mixtures, excess parameter like dielectric constant ($\Delta\acute{\varepsilon}$), dielectric loss ($\Delta\ddot{\varepsilon}$), loss tangent ($\Delta\tan\delta$), activation Energy ($\Delta E_a$) and molar polarization (ΔP_{12}) for binary mixture at 30^0C were reported in Table 2. The feasible associations between AA-solvent and 2-ME-2-ME molecules exhibited different prospective which is completely absent in pure state of liquids. With the aid of excess values, the deviations from ideal behaviour are articulated by different parameters such as dielectric constant and refractive index.

Table-1 Mole fraction (X_A) of Allyl Amine AA and (X_B) 2-ME and its binary mixture, density (ρ), refractive index (n), dielectric constant ($\acute{\varepsilon}$), dielectric loss ($\ddot{\varepsilon}$), loss tangent ($\tan\delta$), activation Energy (E_a) and molar polarization (P_{12})

X_A	X_B	Density ρ	Viscosity η cp	$\acute{\varepsilon}$	$\ddot{\varepsilon}$	Tan ∂	P_{12} cm^3/mole	E_a Kcal/mole	N^2_D
0	1	0.9591	1.52246	9.06152	0.79940	0.08822	57.82596	3.40644	1.97240
0.1317	0.8698	0.9403	1.47475	9.49182	0.90874	0.09574	57.63767	3.38729	2.00070
0.2578	0.7422	0.9157	1.33396	8.47029	0.77287	0.09124	54.93758	3.32687	1.98370
0.3847	0.6153	0.8854	1.18454	8.28683	0.42953	0.05183	54.54411	3.25550	1.99787
0.5103	0.4897	0.8464	1.01818	7.77268	0.76413	0.09831	54.05569	3.16436	1.99795
0.6346	0.3654	0.8302	0.77051	7.93830	0.96538	0.12161	53.75659	2.99650	2.00075
0.7576	0.2424	0.8038	0.6068	7.61249	1.06539	0.13995	52.28085	2.85251	1.31442
0.8794	0.1206	0.7897	0.53120	6.88519	1.01215	0.14700	49.67696	2.77238	2.02069
1	0	0.7610	0.41301	6.38018	1.02394	0.16049	48.45093	2.62083	2.01785

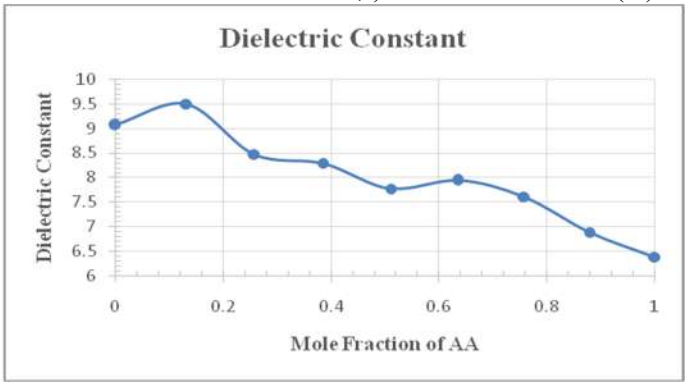
Table-2 Mole fraction (x) of Allyl Amine AA and 2-ME, excess parameter like dielectric constant (Δέ), dielectric loss (Δε̋), loss tangent (Δ tan δ), activation Energy (ΔE_a) and molar polarization (ΔP_{12}) for binary mixture at 30^0 C.

X_A	X_B	Δ η	Δέ	Δε̋	Δtan∂	ΔP_{12}	ΔE_a
0	1	0	0	0	0	0	0
0.1317	0.8698	0.0975	0.7780	0.0793	-0.0021	1.0512	0.0823
0.2578	0.7422	0.1032	0.1000	-0.0844	-0.0156	-0.3942	0.1230
0.3847	0.6153	0.0889	0.2568	-0.4563	-0.0642	0.4401	0.1508
0.5103	0.4897	0.0619	0.0794	-0.1499	-0.0268	1.1669	0.1588
0.6346	0.3654	-0.0480	0.5785	0.0235	-0.0125	2.0704	0.0886
0.7576	0.2424	-0.0824	0.5825	0.0958	-0.0032	1.7847	0.0412
0.8794	0.1206	-0.0192	0.1817	0.0153	-0.0048	0.3591	0.0568
1	0	0	0	0	0	0	0

Result and Discussion

The density, viscosity, molar polarization and activation energy are decreasing with increase in mole fraction of Allyl Amine in the binary mixture. Density of pure AA is less than that of pure 2-ME.The variation of Dielectric constant (έ) with mole fraction (X) of AA in the mixture is depicted in figure1**.** Dielectric constant non-linearly increases with decrease minima by increased mole fraction of AA, which suggest formation of the hydrogen bonded complex.[22]

Figure 1 Variation of Dielectric constant (έ) with mole fraction (X) of AA



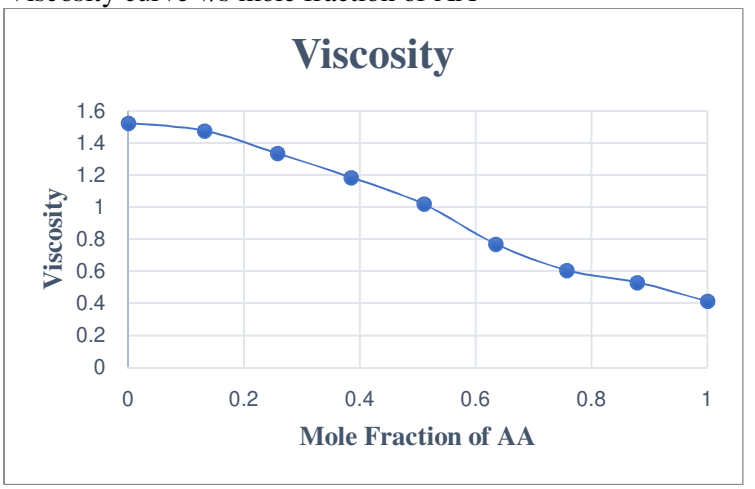
The έ value exhibited in decreasing wave format by increase in AA concentration. The έ values proliferated initially when added AA and reaches to a maximum at approximately X= 0.1317, AA mole fraction with minimum at X= 0.5103, followed by maximum at X= 0.6346 and then declining. The

mixture, ε values are significantly lower compared to the ε values of 2-ME at low AA concentration range, showing stronger hetero-association than the self-association in alcohol molecules. The dielectric constant varies non-linearly as a function of amine concentration in binary systems, due to H-bond interaction between the mixture constituents.

Though Allyl Amine and 2-ME has equal number of carbon atom in their molecular structure, the dielectric constant decreases non-linearly with the increase in the mole fraction of AA. Pure 2-ME has higher dielectric constant. However, R.J. Sengwa (2006)[23] reported that equal number of the carbon atoms in molecular structure of different molecules with hydroxyl groups having high dielectric constant.

According to Figure 2, Viscosity curve indicates that due to solute-solvent interaction, viscosity decreases with increase in the mole fraction of AA[24]. The formation of associates compared with the composition range of the binary mixture is held together by comparatively lesser intermolecular dipole-dipole interactions in the complex.

Figure 2 Viscosity curve v/s mole fraction of AA



The variation of dielectric loss and loss tangent figure (3) and figure (4) with mole fraction of AA shows the 2 maxima (λ_{max}) and single minima is at X=0.3847. In case of 2, 3 DCA + 2EE and aniline + 2-Aloxyethanol, the formation of addact complex is also at the minima. The wobbling nature of the mixture may be due to –O– and –OH group in 2-ME results in multiple complexions[25-27].

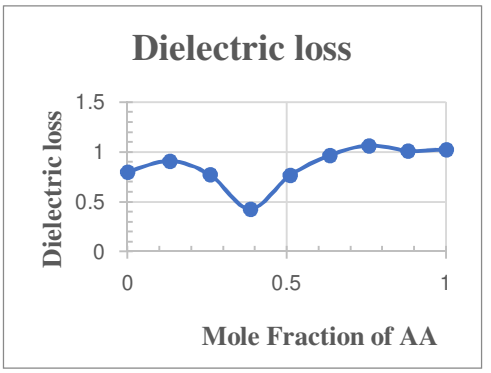


Figure 3 variation of dielectric loss v/s mole fraction of AA

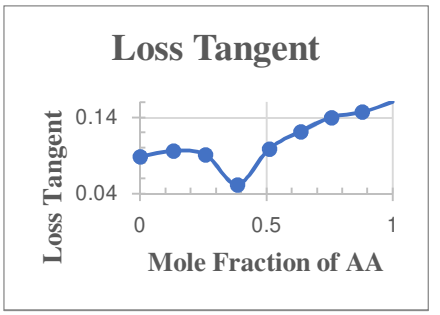
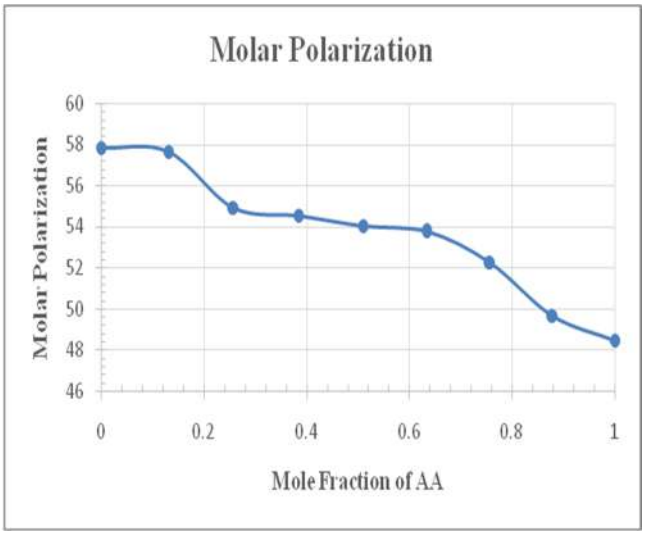


Figure 4 loss tangent v/s mole fraction of AA

Non-linear decrease in molar polarization (figure 5) indicates that the degree of polarity of the solution decreases with increase in mole fraction of AA. Molar polarization is representing 2 regions at high and low concentration. The intersection of the straight line represents separate regions that indicates the point of concentration in which maximum formation of complex at X=0.3847 mole fraction of AA[18]. The variation of excess dielectric constant $\Delta\acute{\varepsilon}$ nonlinearly decreases with minima at X=0.5103**.**

Figure 5 Molar polarization v/s Mole fraction of AA



Excess Parameters

The excess dielectric constant Δέ , excess dielectric dielectric loss (Δε̋),, loss tangent (Δ tan δ), excess activation Energy (ΔEa) and excess molar polarization (ΔP12) values of pure and binary mixtures of AA and 2-ME are calculated for various mole fractions at room temperatures. The excess dielectric constant Δέ indicates the strength and nature of intermolecular interactions in binary liquid mixtures. Observation of variation of the excess dielectric constant (Δέ) of AA with 2ME binary mixtures are positive and less in magnitude. According to Ch. V. V. Ramana [28], dielectric constant (Δέ) > 0 indicates that there is an increase in number of effective dipoles contributed in the mixture dielectric polarization, results in formation multimers and dimmers .The positive excess dielectric constant (Δέ), figure (6) also suggest that the effective number of dipoles in the mixture might be greater than the corresponding average number in the pure liquids, probably due to the creation of new structure leading to a higher macroscopic permittivity [29,30]

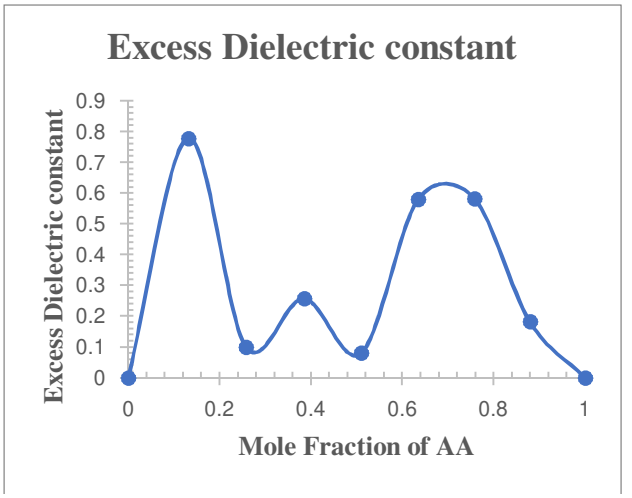


Figure 6 The variation of excess dielectric constant Δέ with mole fraction of AA+2-ME

The variation of excess dielectric loss (Δε̋) is observed in figure 7 showing negative for the low molar concentration region and positive for the high molar concentration region of AA showing minima at X= 0.3847. The dielectric loss is regarded due to molecular motion which is governed by the complex forces of molecular interactions. Variation in excess loss tangent figure 8 supports this showing minima at X=0.3847 and is negative for entire molar concentration region of AA.

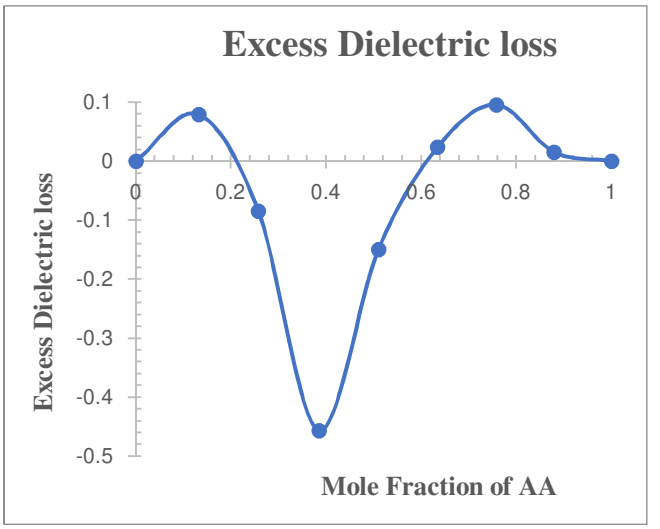


Figure 7 Excess dielectric loss (Δε̈) with mole fraction of AA+2-ME

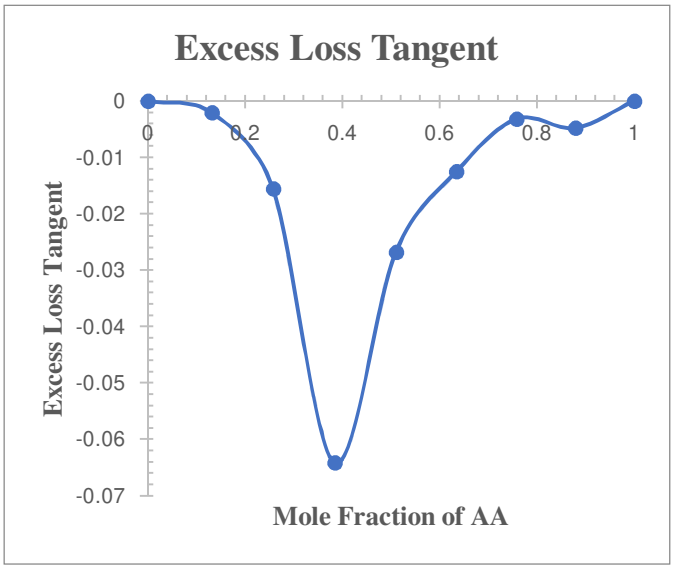


Figure 8 Excess loss tangents with mole fraction of AA+2-ME

According to Solimo and Riggio[31] positive values of excess viscosity due to strong specific interaction causes complex formation and negative value of

excess viscosity are observed for system of different molecular size in which the dispersion forces are dominant.

Excess viscosity figure 9, is initially positive upto X=0.5103 and later on becomes negative as the mole fraction AA increases .This may be due to strong specific interaction and system of different molecular size in which the dispersion forces are dominant giving an estimation of the intermolecular interaction[32].

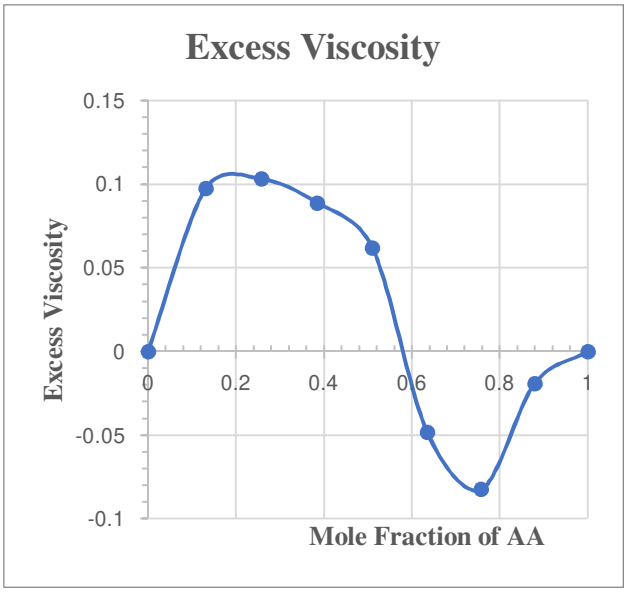


Figure 9 Excess viscosity v/s mole fraction AA+2-ME

Excess Activation energy ΔE_a, noted in figure 10 is positive indicating strong interaction between AA and 2-ME molecules. Maxima occur at about X=0.3847 and X= 0.5103 Δέ designates that there is a strong interaction between solute and solvent[9, 33]. The similar was observation were noted in case of propane diol (PD) and Etyhylene diamine (EDA) molecules[32].

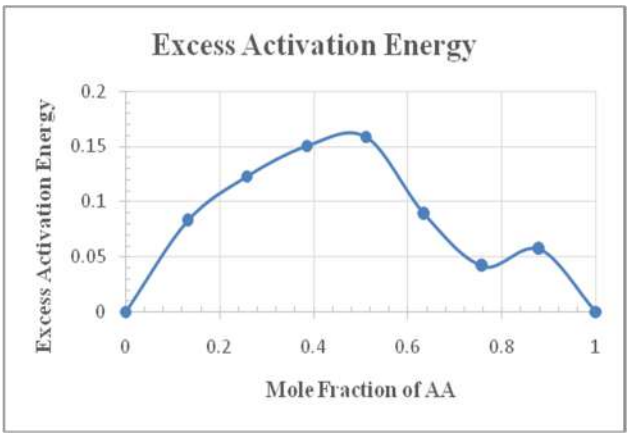
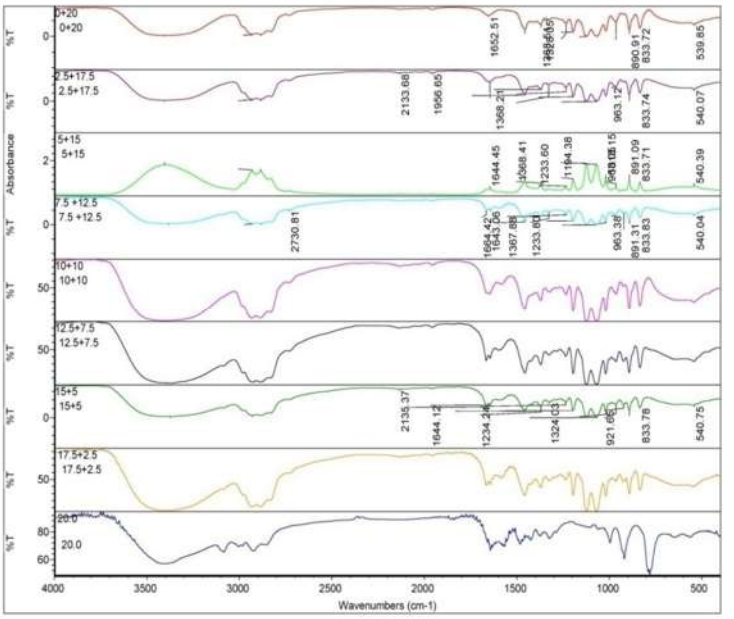


Figure 10 The Plot of excess activation energy (ΔEa) with mole fraction of AA+2-ME

FT-IR Spectral Analysis

The figure11 shows the FT-IR spectra of AA in 2-ME at room temperature between 3000 cm-1 and 3800 cm-1, in the range of the hydroxyl stretching vibration, these spectra strongly influenced by H-bonding. Small variation in frequency shift is observed indicating solute – solvent interaction. No significant change in the hydroxyl group bands with various concentrations, the spectrum is a strong indication of intra molecular (internal) hydrogen bonding[34]. Lump at 2730.81cm^{-1} gradually disappeared at X=0.3847 indicating complex formation. In the absence of 2-ME, characteristic carbonyl band has been observed at 1652 cm^{-1} by increasing concentration of 2-ME decreases the intensity of the original band. The bands due to linear bonded methoxy CO is observed in the 2000–2170 cm−1.The bands observed at 1956 cm-1is due to the formation of intermolecular bond between AA and 2ME. New band appear at the frequency of 1643.08cm $^{-1}$when the concentration of AA is higher. This behavior indicates the existence of 1:1 and 1:2 complexes[35], observed the same behavior of p-chlorophenol in carbon tetrachloride at 25^{o}C.Characteristic carbonyl band has been observed at 1664-1641 cm^{-1} that is in the range of 1500 and 1750 cm^{-1} of the carbonyl stretching vibration. The Fourier-transform infrared spectroscopy (FTIR) analysis of AA+2-ME proved the presence of amines, alcohols, Nitro compounds, carboxylic acids, esters, ethers and hydroxyl group. The spectrum clearly indicates multiple functionality, occurring mutual interaction.[36]

Figure 11 FTIR Spectra of AA and 2-ME at various concentration (0+20, 2.5+17.5, 5+15, 7.5+12.5, 10+10, 12.5+7.5, 15+5, 17.5+2.5 and 20+0)



Conclusions

Values of dielectric parameter, viscosity, activation energy have been reported for different mole fractions of AA. Dielectric loss, loss tangent, excess parameters like excess dielectric loss ($\Delta\varepsilon$) , excess loss tangent suggests possibility of complex formation at X=0.3847 and X= 0.5103 which is confirmed by the molar polarization curve. Excess activation energy indicates strong interaction between unlike molecules. The molar polarization curve and FTIR suggests approximately 1:2 type complex formations in the mixture. This positive excess permittivity of the mixtures also suggests any significant intermolecular interaction is effectively present at room temperature.

The FTIR spectra of binary mixture have been recorded and analyzed which clearly indicates multiple functionality, occurring mutual interaction between AA and 2-ME.The above evaluation is advances in biomedical, pharmaceutical and industrial application.

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Declaration of interest statement:

We wish to confirm that there are no known conflicts of interest associated with

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