ACOUSTIC PARAMETER INVESTIGATION OF TERNARY MIXTURE OF N-BUTANOL, WATER AND ACETIC ACID BY USING ULTRASONIC TECHNIQUE

A

Dissertation Submitted By

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In

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Under the guidance of

Dr. K. C. Juglan

(Supervisor)

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CERTIFICATE

This is to certify that Akanksha Dixit has completed her M.Sc. Dissertation entitled **"ACOUSTIC PARAMETER INVESTIGATION OF TERNARY MIXTURE OF N-BUTANOL, WATER AND ACETIC ACID BY USING ULTRASONIC TECHNIQUE"** under my guidance and supervision. To the best of my knowledge, the present work is the result of her original investigation and study. No part of the dissertation has ever been submitted for any other degree or diploma at any other University.

The dissertation is fit for the submission and the partial fulfillment of the conditions for the award of degree of **M.Sc. (Hons) Physics**.

Dr. Kailash Chandra Juglan **Dated:**

COD, Department of Physics,

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Phagwara (Punjab)

DECLARATION

I do hereby state and declare that the dissertation submitted for the partial fulfillment of the requirement for the degree of **Master of Science** in **Physics (Hons.)** entitled as **"ACOUSTIC PARAMETER INVESTIGATION OF TERNARY MIXTURE OF N-BUTANOL, WATER AND ACETIC ACID BY USING ULTRASONIC TECHNIQUE"** is entirely my original work, and all ideas and references used have been duly acknowledged. This work is not submitted for award of any other degree or diploma at any other University or Institution.

Dated:

Akanksha Dixit

Regd. No. 11203853

DEDICATED TO

MY LOVING PARENTS

&

MY BROTHER

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Akanksha Dixit

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SYMBOLS

U	Ultrasonic Velocity of wave
f	Frequency of particle vibration
λ	Wavelength of wave
η_1	Viscosity of water
η_2	Viscosity of experimental liquid
ρ_1	Density of water
ρ_2	Density of experimental liquid
t_1	Time of flow of water
t_2	Time of flow of experimental liquid
W_1	Weight of distilled water
W_2	Weight of experimental liquid
β	Adiabatic compressibility
Z	Acoustic impedance
L_f	Intermolecular free length
α/f^2	Ultrasonic attenuation
τ	Relaxation time
M	Effective molecular weight
V	Available free volume
K	Temperature independent constant
V_m	Molar volume
V_a	Availabe volume
W	Wada's constant
R	Rao's Constant
b	Vander waal's constant
P	Internal pressure
ΔG	Gibb's free energy
K_B	Boltzmann's Constant
T	Absolute temperature
h	Planck's constant
H	Enthalpy

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ABTRACT

Ultrasonic velocity, Density and Viscosity ofternary mixtures of n-Butanol, Water and Acetic acid ($C_4H_9OH+H_2O+CH_3COOH$) have been measured at a constant temperature 289K at different concentrations by using Ultrasonic Technique. From experimentally obtained data other Acoustical parameters like Adiabatic Compressibility, Adiabatic Impedance, Intermolecular Free Length, Available Free Volume, Relaxation Time, Ultrasonic Attenuation, Wada's constant, Rao's constant, Vander Waals constant, Internal Pressure, Gibb's Free Energy and Enthalpy have been calculated. In this non-ideal mixture, nonlinear variation of acoustical parameter in the composition of the chemical system suggests the presence of weak molecular interaction.

CHAPTER 1

Introduction:

Sound is transmitted through a medium by inducing vibrational motion of the molecules through which it is traveling. This vibrational motion represents the sound frequency [6]. Ultrasound is sound of a frequency that is above the threshold of human hearing. The lowest audible frequency for humans is about 18Hz and the highest is normally around 18-20 kHz for adults, above which it becomes inaudible and is defined as ultrasound [6]. Some other living creatures like bats can hear these kinds of waves. The returning echo from an object situating apart from the bats help them to find the obstacles in the path.

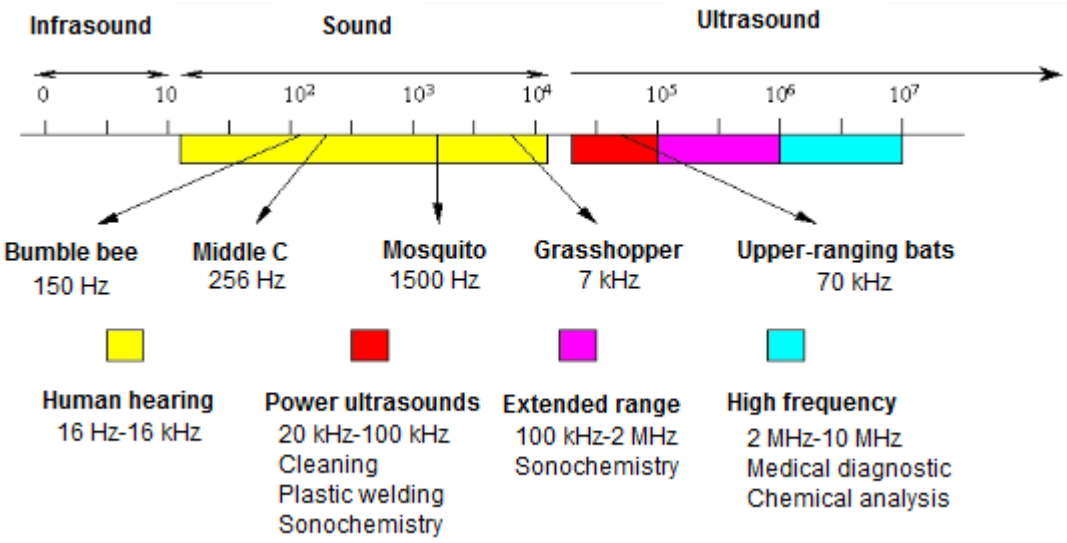

Figure1.1:http://www.intechopen.com/source/html/41410/media/image1.png

Starting from the basics, classical principles of sound waves established by Rayleigh's Theory of Sound and Lamb's Dynamical Theory of Sound to the present ongoing series of theories incorporated with their applications and extensions of basic principles have evolved. The ultrasonic is progressive and periodical appearance of an era that has the basic connection to piezoelectricity and magnetostriction as a source of

generating acoustic waves in media [22]. Also, it is further related with transduction. The ultrasonic method encompasses the analysis, examining and processing of techniques related to the vibrational energies. These ultrasonic waves are of very high amplitude and high frequencies. An ultrasonic wave travels with a constant speed in a homogeneous medium. These waves show reflection, refraction, attenuation and scattering phenomena similarly as light wave does.

In case of non-homogeneous medium the defects like cracks, impurities or different densities in the medium can be found because the amplitude of the wave along with its intensity changes (or decreases). Due to all these properties, ultrasonic waves are very useful nowadays. The technical prospectus of the ultrasonic technology has a vast variety of devices and instrumentations.The applications that use low amplitude waves include Pulse Coding Devices; Waveform Filtering and many Non-destructive testing types of equipment used in determining defects in metals, glasses etc. Surface waves are implemented in RADAR, SONAR and Communication Systems. High amplitude waves are used in cavitation effects for chemical and metallurgical control of material texture [22]. These waves are used to measure the distances, depth of sea, to heat up the matters, to accelerate chemical reactions, to solder aluminium, to sterilize the water & milk, to mix the alloys of different compositions etc. The next important use lies in the diagnostics and therapeutics in medical field to detect tumors & cancer, in relieving neuralgic pains, in bloodless surgery, in imaging for reproducing pictures by holographic means and nondestructive evaluation of surfaces of body and inner organsand in sonography also.

The main purpose of using this technique in chemical analysis is its non-destructive kind of nature (NDT). Other such methods include magnetic-particle, radiographic, liquid penetrate, low coherence interferometry and eddy-current testing. Among all these methods, ultrasonic technique has been found to be most powerful tool in the investigation of structure and molecular interactions occurring in the liquid mixtures. Also it gives valuable information about the physical properties and strength of molecular interactions in the mixtures[14].This method can also detect internal flaws inside the substance and cracks on the surfaces.

The ultrasonic technique provides vast information about inter-molecular and intra-molecular interactions in different types of chemical mixtures. These interactions depend upon the combination of chemicals used to form a system and are of different types like dipole-dipole interaction (in polar-polar system), dipole-induced dipole interaction (in polar-non polar system), ion-dipole interaction, etc. Theintermolecular interactions influence the structural arrangement along with the shape of the molecules and further interpret the nature and behavior of the system.

In ultrasonic techniques the acoustic waves are produced by piezoelectric generator. When some pressure or compression is subjected on two opposite faces of a quartz crystal (piezoelectric crystal), generation of charges take place on a set of opposite faces which are perpendicular to the faces at which pressure is applied. The magnitude of charge produced is directly proportional to the amount of pressure applied to the surfaces. Charge developed on one face is positive and on other face is negative. Furthermore, if instead of compression, the faces of crystal are subjected to some tension, the nature of charges generated also gets reversed. The process of appearance of charges on transverse faces of certain crystal when subjected to external stress is called Piezoelectric Effect.

But if an electric field is applied across two opposite faces of a quartz crystal, then extension or compression is produced in the crystal in a direction transverse to the direction of electric field. This effect is called Anti-piezoelectric effect. The extent of compression or rarefaction is proportional to the strength of the electric field applied.

If the applied electric field is alternating in nature, then quartz crystal starts vibrating at the frequency of the electric field and hence produces acoustic waves in the air. If the frequency of the electric field is same as the natural frequency of the crystal, then the amplitude of the crystal oscillations is quite large. This property of quartz crystal is used for producing ultrasonic waves. Apart from quartz, other materials showing this property are Rochella Salt (Sodium potassium tartrate), Tourmaline etc. Some other synthetic ceramic materials that possess piezoelectricity property are Barium Titanate and Lead Zirconium Titanate (PZT).

Distributed Point Source Method:

Distributed Point Source method (DPSM) is a semi-analytical technique used to model ultrasonic (or eddy current) transducers for computing pressure and velocity fields (or magnetic fields) generated by it. In the DPSM technique, point sources are placed close to the active front face of the transducer and the target boundary, as well as on both sides of an interface. Each point source acts as an active source of energy and for each of them an analytical solution for the produced field can be obtained. The total ultrasonic field is calculated by superimposing the solutions of all point sources. In order to correctly calculate the total ultrasonic field, it should be kept in mind that the second transducer not only acts as a generator of the ultrasonic energy, but also as a scatter of the energy generated by the first transducer, and vice versa [8].

This scattering effect or "interaction effect" must be considered in order to correctly model the complete ultrasonic field. It is interesting to see that the interaction effect reduces the total field strength on many occasions. When the transducers are placed at a larger inclination, relative to each other, the interaction phenomenon affects the ultrasonic field less in comparison to the case when they are placed at a smaller inclination angle [8].

CHAPTER 2

Review of literature:

Thiyagarajan et al. (2007) [28] measured acoustical parameters at 303K in ternary mixture of aniline, methanol and cyclohexane. This type of study related with the concentrations is useful in getting information about structure and bonding of associated molecular complexes. Ultrasonic velocity, density and viscosity increases monotonically but non-linearly. This non-linear variation specifies the presence of intermolecular interactions. Dipole-dipole interactions are formed between the hydrophilic group (OH) of ethanol and the amino group of aniline whereas weak dispersive interactions are formed between the cyclohexane rings and the hydrophobic group of ethanol.

Mehra et al. (2007) [9] studied the molecular interactions in binary mixtures of benzene-butanol and toluene-butanol systems by measuring acoustical parameters along with thermo dynamical parameters at various temperatures. All these parameters show large variation with respect to the mole fraction of benzene and toluene at all temperatures. The low magnitude of excess molar volume is the indication of weak dispersion forces or dipole-induced dipole type of forces of weak magnitude present between the components of the system. The addition of non-polar benzene and toluene in 1-butanol leads to depolymerization (of 1-butanol) by the breaking of hydrogen bonds.

Thirumaran et al. (2008) [25] investigated acoustic parameters like ultrasonic velocity, density and viscosity for four amino acids L-alanine, L-leucine, L-valine and L-proline in aqueous sodium acetate solution at temperatures 303K, 308K and 313K. In all amino acid systems the value of density, viscosity and ultrasonic velocity increases with increase of molar concentration of amino acids. For the same combination, these parameters decrease with increase in temperature, except ultrasonic velocity. The work was concluded that the strong molecular association is found in L-leucine system than the other three amino acids and hence L-leucine is acting as effective structure maker than other amino acids.

Singh et al. (2009) [19] computed first-, second- and third-order elastic constants, ultrasonic velocity, gruneisen parameters, non-linearity parameter, debay temperature and thermal relaxation time. Thermo elastic losses are compared to phonon-phonon

interaction of intermetallic compounds and further the theoretical and experimental results are analyzed together. Chemicals used are AgMg, CuZr, AuMg, AuTi, AuMn, AuZn and AuCd having CsCl type structure (B2 structure).

Zorebski et al. (2010) [29] measured ultrasonic velocity and densities in binary mixture of 1, 2-ethanediol with 2-ethyl-1-hexanol, 1-heptanol and ethanol for different compositions at temperature 298.15K by using pulse-echo-overlap method and vibrating-tube densimeter. The negative values of the excess molar volumes and isentropic compressibility over the entire composition range are observed in the case of ethanol; whereas in case of 2-ethyl-1-hexanol, 1-heptanol and 1-nonanol, the positive values of the excess molar volumes are observed over the entire composition range.

Ravichandran et al. (2010) [17] measured acoustical parameters in the liquid mixture of zinc sulfate and zinc nitrate at temperature 303K. They show variation with the change of mole fraction of the system of chemicals. The increase in concentration of Zinc nitrate weakens the molecular forces and hence there comes a change in velocity. This non-linear variation of velocity with increase in concentration indicates the complex formation between the constituents of a mixture. Also the results were interpreted in the terms of ion-dipole interaction in the mixture.

Singh and Bhatt (2010) [20] measured ultrasonic velocity, viscosity and density of the solution of polyvinyl acetate in acetic acid at different concentrations 0.50% to 2.00% and at temperature range 35℃ to 55℃. By using measured parameters other acoustical parameters are calculated like adiabatic compressibility, acoustic impedance, relaxation time and attenuation. Variation of these parameters with temperature and concentration is studied. This provides an appropriate understanding about the strong solute-solvent interaction in polymer solution at higher concentration. Variation in Ultrasonic velocity signifies structural re-adjustment of molecular packing in the solution. Decrease in adiabatic compressibility with increase in concentration indicates the enhancement of the bond strength at that particular concentration.

Pavai and Renuka (2011) [11] examined ternary chemical mixture or propyl acetate, cyclohexane and 2-methoxyethanol by taking different compositions at 303K,

308K and 313K. By using ultrasonic technique, ultrasonic velocity, density and viscosity were measured and other parameters as adiabatic compressibility, inter molecular free length, free volume, internal pressure, acoustic impedance, Gibb's free energy and their excess values have calculated. The adiabatic compressibility and free length increases with mole fraction of propyl acetate and it signifies weakening of hydrogen bond between propyl acetate and 2-methoxyethanol. Decrease in Gibb's free length with increase in concentration of propyl acetate suggests that the formation of activated species that in necessary for the viscous flow is easier in propyl acetate rich region. The temperature dependence of the different parameters suggests that the degree of deviation from ideality is temperature sensitive and influenced by the weak hydrogen bond.

Chimankar et al. (2011) [2] studied the nature and strength of molecular interaction in mixture of lysine and Adenosine with water. Ultrasonic velocity, viscosity and density were measured and other parameters such as adiabatic compressibility, relaxation time, Rao's constant, Wada's constant, Vander Waal's constant, Acoustic impedance, free length, internal pressure had calculated. Non-linear variation of ultrasonic velocity, relaxation time and other thermal parameters with concentration of Lysine and Adenosine with water signifies the complex formation in the mixture. Increase in hydration helps to increase hydrogen bonding with increase in concentration. Also, it provides increase in hypochromacity which further indicates stronger solute-solvent interaction with higher concentrations.

Chimankar et al. (2011) [3] measured ultrasonic velocity and density of ternary mixture of Glycylglycine, Sodium Chloride and water by using PEO technique and hydrostatic plunger method. Using this data other thermodynamic parameters like molar volume, volume expansivity, specific heat, molecular radius, Molwyn-Huhges parameter, cohesive energy density, internal pressure, change in entropy, disorder parameter, interaction energy and solubility parameter were calculated using standard formulae. The presence of solute-solvent interactions through dipole-dipole interaction of –OH groups of peptides with the surrounding water molecules. Increase in specific heat with increase in concentration indicates structure making ability of di-peptide i.e. hydrophobic

hydration or structure enhancing property of dipeptide with a rise in concentration of the dipeptide.

Thirumaran and Sathish (2011) [26] studied the structure making and structure breaking behavior of divalent metal sulphates in aqueous Ethylene Glycol at 308.15, 313.15 and 318.15K. The ternary mixture of water, ethylene glycol and metal sulphates (manganese sulphate, nickel sulphate and cobalt sulphate) is taken for the measurement of fundamental acoustical parameters like ultrasonic velocity, viscosity and density. The transfer volume studies which which predict the solute-co-solute interactions suggesting that ionic-hydrophobic interaction are existing in all three metal sulphates systems. Strong inter-ionic interactions like solute-solvent, ion-solvent interactions are existing; however a weak ion-ion interaction is noticed. Molecular interaction follows the order: manganese sulphate > nickel sulphate > cobalt sulphate.

Palani et al. (2011) [10] measured ultrasonic velocity, viscosity and density of L-histidine, L-arginine and L-lysine in aqueous sucrose (0.5M) solution at 298, 303 and 308K. Other parameters as adiabatic compressibility, apparent molal volume, limiting apparent molal compressibility, associated constant, hydration number and the values of A and B coefficients of the Jones-Dole equation were computed. All these parameters provide a significant ways to describe intermolecular interactions. Positive value of hydration number suggests that the compressibility of the solution will be less than that of the pure solvent. So solute gains mobility and have a greater probability of solvent molecules to come closer to adjacent molecules. Magnitude of apparent molal volume shows the order L-histidine > L-arginie > L-Lysine. It concludes that L-lysine is a more effective structure maker than other amino acid usd in this particular experiment.

Sastry et al. (2012) [18] used ultrasonic technique to investigate the molecular interaction in pure liquid-liquid mixture and ionic interactions in electrolytic solutions. This interaction was carried out by the measurement of ultrasonic velocity, density and viscosity by using the standard apparatus. The molecules of component chemicals were forming hydrogen bonding through dipole-dipole interaction. The ultrasonic velocity varies with concentration due to solute-solvent interaction followed by molecular association.

Thabane et al. (2012) [24] studied that the variation of acoustical parameters (ultrasonic velocity, density and viscosity) shows non-linear relation with molar concentration. These parameters also give an idea about the nature and strength of molecular interactions present in the system. A thermoplastic polymer "Polyvinyl chloride" is used along with tetrahydrofuran (as a solvent) as an experimental mixture at different concentrations. Conclusion was made that ultrasonic velocity is related non-linearly with concentration and the increase in internal pressure with increase in concentration of polyvinyl chloride explains the attractive forces between the solvent and polymer molecules.

Rani et al. (2012) [16] studied solute-solvent interactions using acoustic and spectroscopy techniques. Derivatives of L-Arginine in non-aqueous solution is taken as the sample and fundamental parameters, namely ultrasonic velocity, density and viscosity are determined for different concentrations ant at different temperatures. Other thermodynamic parameters are computed and analyzed with the help of these fundamental parameters. This study suggests that the intermolecular hydrogen bonding exists in the amino acid moiety. This particular mixture exhibits in non-zwitter ionic (charge solvated) form. Sample is in di-ionic form. From the spectral analysis salvation with solvent molecules occurs at NH^+ site of guanidine group. The results obtained from acoustic and thermodynamic analysis are well correlated with spectroscopic study.

Thirunavukkarasu and Kanagathara (2012) [27] measured ultrasonic velocity, viscosity and density to investigate the behavior of Toluene and Carbon Tetra Chloride at various concentrations at different temperatures. Other acoustical parameters like adiabatic compressibility, intermolecular free length, Rao's constant, internal pressure and free volume were calculated to predict the nature and strength of molecular interaction between the solute (Toluene) and solvent (Carbon Tetra Chloride). At lower concentrations interaction are very less but at higher concentrations the solute molecules are pushed closer to the solvent molecules, there by producing hydrogen bonds. Variation in adiabatic compressibility and free length indicates the possibility of complex formation like hydrogen bond.

Praharaj et al. (2013) [13] measured fundamental acoustical parameters for the ternary chemical mixture of N,N-dimethylfomamide, Cyclohexane and Benzene at 288, 298, 308 and 318K. Other excess parameters were calculated with the help of these parameters. Ultrasonic velocity, viscosity and density decrease with increase in mole fraction on DMF. This is due to the dipole-induced dipole interaction between DMF and other two non-polar components. They show a reverse trend when the temperature increases. Parameters like Gibbs' free energy, acoustic impedance, relaxation time also indicate a weak dipole-induced dipole interaction between the components, which is confirmed by the nature of excess values.

Srinivasu et al. (2013) [22] have investigated the non-ideal behavior of some physico-chemical liquid mixtures by measuring the values of ultrasonic velocity, density and viscosity. The mixture was the binary combination of 1-butanol and hexane at different temperatures 313.15K, 318.15K and 323.15K. Thermodynamic parameters like adiabatic compressibility (β), acoustic impedance (z), molar volume (V_m) etc are calculated and nature of molecular interaction was studied. Ultrasonic velocity and adiabatic compressibility, independent of temperature and pressure, determine the orientation of the solvent molecules around the liquid molecules. Adiabatic compressibility and free length is inversely proportional to the ultrasonic velocity, and so as the relation of these parameters towards the mole fraction. Also the acoustic impedance increases with increase in mole fraction of 1-butanol.

Wadekar (2013) [29] examined Schiff base (E)-2-(2-chlorobezylideneamino) benzoic acid and its metal complex of Fe (III) in DMSO. Ultrasonic velocity, viscosity and density were calculated. Other acoustical parameters and their excess values are computed and presented as function of compositions. Ultrasonic investigations of liquid mixtures consisting of polar and noon-polar components are of considerable importance in understanding intermolecular interactions between the component molecules. Increase in viscosity with increasing concentration suggests a strong solute-solvent interaction. A positive deviation of internal free length and negative deviation in compressibility indicates that the molecules are nearer in the system. Other values signify that system is stabilized in greater extent.

Singh et al. (2013) [21] studied acoustical parameters namely ultrasonic velocity, isentropic compressibility, acoustic impedance and intermolecular free length due to the importance and utility of ethyl methyl ketone as a nuclear extract used in atomic energy industry. The excess parameters β^z, Z^E were fined for different mole fractions of ethyl methyl ketone. The ultrasonic velocity increases with increase concentration of ethyl methyl ketone. The conclusion for the whole experimental and theoretical study is the formation of micro heterogeneous clusters of unlike molecules.

Dash et al. (2013) [5] made a study on ternary mixture of dimethyl acetamide (DAMC) in diethyl ether and acetone by measuring acoustical parameters at different frequencies (2MHz, 4MHz, 6MHz, 8MHz) at temperature 308K. Other thermo acoustic parameters were computed by using different relations between all these parameters. Multi-frequency interferometer is used to measure the ultrasonic velocity. The study was concluded that molecular interaction decreases with the increase in the frequency for fixed concentration of DAMC in this particular mixture.

Kaur and Juglan (2013) [7] studied ultrasonic velocity, density and viscosity of binary mixture of Polyvinyl Acetate and Acetic Acid at temperature 299 K and by taking different concentrations of both the components. Measured acoustical parameters are used to calculate other thermo acoustical parameters. Presence of solute-solvent interaction is confirmed by the trend of viscosity. Also it suggests that there is more association between solute and solvent molecules. Linearity of Rao's and Wada's constants in the plots drawn with respect to concentration signifies the absence of complex formation in chemical system.

Bhandakkar et al. (2014) [1] investigated acoustical and excess thermodynamic parameters of binary mixture of Cinnamaldehyde and liquid acetone. These parameters shed light on the solute-solvent and dipole-dipole interactions. Also the variation of parameters was a function of temperature is non-linear and this further interprets the presence of solute-solvent, solvent-solvent and dipole-dipole interaction. For the observed molecular interaction, hydrogen bond formations are responsible for the heteromolecular interaction in the liquid mixture used. Excess transport properties of the binary liquid mixture are considered to be a reflecting agent of magnitude of polarity and

size of the molecules in the interaction. The strength of interaction tends to weaker which may due to the presence of weak intermolecular forces and thermal dispersion forces.

CHAPTER 3

Scope of study:

The present study has been done to measure the ultrasonic velocity and other acoustical parameters for the ternary mixture of n-Butanol, Acetic Acid and Water. This mixture is widely used as a running solvent in TLC for Peptides, Amino Acids and many other chemicals and also to monitor the progress of a reaction. Different concentrations are used for different chemical mixtures. The behavior of parameters with variation in concentration will help to find out the most interacting ratio of concentrations and furthermore it will optimize the efficiency of work.

CHAPTER 4

Methodology:

4.1 Objective:

To investigate the acoustical parameters, i.e. ultrasonic velocity, density, viscosity & other thermodynamic parameters of chemical mixture containing n-butanol, water and acetic acid at different concentrations of each constituent.

4.2 Research Methodology:

n-butanol of Assay (GC) grade of molecular weight 74.12 gm/mol, Acetic Acid of Assay (acidimetric) of molecular weight 60.05gm/mol and Distilled Water is used.

For the measurement of ultrasonic velocity ultrasonic interferometer (Mittal enterprises-F-80 model) at a standard frequency of 2 MHz with an accuracy of ± 0.1 m/s is used.The density of the mixture is measured with the help of "Specific gravity bottle" whose capacity is 25ml and the viscosity of the mixture can be measured accurately by "Oswald's Viscometer" whose capacity is 10ml with a precision of ± 0.1kgm^{-3}and 0.1% respectively. Distilled water has been used as a standard liquid with time flow been measured with an accuracy of 0.1 sec. The whole experiment has been done at temperature 289 K.

Other thermo-dynamical parameters such as adiabatic compressibility, adiabatic impedance, intermolecular free length, available free volume, relaxation time, ultrasonic attenuation, effective molecular weight, free volume, molar volume, available volume, Gibb's free energy, enthalpy, Wada's constant, Rao's constant, Vander Waal's constant and internal pressure will be calculated with the help of standard parameters.

4.2 Measurement of standard acoustical parameters:

1. Ultrasonic Velocity:

Ultrasonic velocity (U) can be given as,

$$U = \lambda \times f$$

Where U = velocity of ultrasonic wave

λ = wavelength of the wave

f = frequency of particle vibration

2. Viscosity:

To measure the viscosity of the chemical mixture Ostwald's Viscometer is used. By obtained data viscosity can be calculated using the following formula,

$$\eta_2 = \eta_1 \frac{t2}{t1} \frac{\rho 2}{\rho 1}$$

Where η_1 = Viscosity of water

t_1 = Time of flow of water

ρ_1 = Density of water

η_2 = Viscosity of the mixture

t_2 = Time of flow of the experimental liquid

ρ_2 = Density of the experimental liquid

3. Density:

Density of the chemical mixture can be calculated by using specific gravity bottle. Relation for this can be written as,

$$\rho_2 = \rho_1 \frac{W_2}{W_1}$$

Where W_1 = Weight of the distilled water

W_2 = Weight of the experimental liquid

ρ_1 = Density of water

ρ_2 = Density of experimental liquid

4.3 Measurement of other acoustical parameters:

1. Adiabatic compressibility:

The adiabatic compressibility is the measure of relative volume change of a fluid or solid to as a response of pressure change and can be given as,

$$\beta = \frac{1}{\rho U^2}$$

Where β = Adiabatic compressibility

U = Ultrasonic velocity

ρ = Density of medium

2. Acoustic Impedance:

Acoustic impedance is the opposition offered to the flow of sound through the surface of medium and can be expressed as,

$$Z = \rho \times U$$

Where Z = Acoustic impedance

ρ = Density of medium

U = Ultrasonic velocity

3. Intermolecular free length:

Intermolecular free length is the distance between surfaces of two neighboring molecules and can be written as,

$$L_f = K\,(\beta)^{1/2}$$

Where L_f = Intermolecular free length

K = Temperature dependent constant or Jacobson's constant

β = Adiabatic compressibility

4. Ultrasonic attenuation:

When an ultrasonic wave propagates in a medium there is a decrease of energy in wave depending upon the properties of medium, it is known as ultrasonic attenuation. It can be written as,

$$\frac{\alpha}{f^2} = \frac{8\pi^2\eta}{3\rho U^2}$$

Where η = Viscosity of mixture

ρ = Density of mixture

U = Ultrasonic velocity

5. Relaxation time:

Relaxation time is a measure of the time taken by medium particles to come back to their mean position within medium following displacement by ultrasonic wave. It can be expressed in the following manner,

$$\tau = \frac{4\beta\eta}{3}$$

Where τ = Relaxation time

β = Adiabatic compressibility

η = Viscosity of mixture

6. Effective molecular weight:

Effective molecular weight is the algebraic sum of the product of the mole fraction and molecular mass of each constituent used and can be written as,

$$M = \sum_{i=1}^{n} [m_i x_i]$$

Where, m_i= Molecular mass of constituent

x_i= Mole fraction of that constituent

7. Available free volume:

It is the free space between the molecules to move and can be given as,

$$V = \left[\frac{MU}{K\eta}\right]^{3/2}$$

Where V = Free volume

M = Effective molecular weight

U = Ultrasonic velocity

K = Temperature independent constant

η = Viscosity of mixture

8. Molar volume:

The molar volume is the volume occupied by one mole of a substance (chemical element or chemical compound) at a given temperature and pressure. It can be defined as,

$$V_m = \frac{M}{\rho}$$

Where, Vm= Molar Volume

M= Effective molecular weight

ρ = Density of chemical mixture

9. Available Volume:

Available Volume is a direct measure of the compactness and strength of bonding between the molecules of the system and can be defined as,

$$V_a = \frac{M}{\rho}\left(1 - \frac{U}{U_\infty}\right)$$

Where, V_a= Available volume

M= Effective molecular weight

U= Ultrasonic velocity

U_∞= Schaaf's limiting value 1600 m/s for liquids

10. Wada's constant:

It is a relation between adiabatic compressibility, effective mass and density of the mixture. Relation is as follows,

$$W = \frac{M}{\rho\sqrt{\beta}}$$

Where W = Wada's constant (independent of temperature)

β = Adiabatic compressibility

M = Effective molecular weight

ρ = Density of mixture

11. Rao's constant:

It is a simple relation between velocity of ultrasound and density of mixture and can be given as,

$$R = \frac{MU^{1/3}}{\rho}$$

Where R = Rao's constant

U = Ultrasonic velocity

M = Effective molecular weight

ρ = Density of mixture

12. Vander Waals' constant:

It can be explained as volume excluded by a mole of particles. It can be expressed as,

$$b = V\left[1 - \left(\frac{RT}{MU^2}\right)\left\{\left(1 + \frac{MU^2}{3RT}\right)^{\frac{1}{2}} - 1\right\}\right]$$

Where b = Vander Waals' constant

R = Gas constant

T = Room temperature

M = Effective molecular weight

U = Ultrasonic velocity

13. Internal pressure:

Internal pressure is a measure of how the internal energy of a system changes when it expands or contracts at constant temperature. Relation for internal pressure is given by,

$$P = bRT\left[\left(\frac{K\eta}{U}\right)^{1/2}\left(\frac{\rho^{2/3}}{M^{7/6}}\right)\right]$$

Where P = Internal pressure of mixture

b = Vander Waals' constant

R = Gas constant

T = Absolute temperature (room temperature)

14. Gibb's free energy:

Gibbs free energy is the measure of the process-initiating work obtainable from a system at a constant temperature and pressure. It can be defined as,

$$\Delta G = K_B T \ln\left[\frac{K_B T \tau}{h}\right]$$

Where, ΔG = Gibb's free energy

K_B = Boltzmann's constant

T = Absolute temperature

τ = Relaxation time

h = Planck's constant

15. Enthalpy:

Enthalpy of a solution is defined as the overall amount of energy which is released or absorbed during the interactions between the solutes or/and solvents. It can be written as,

$$H = V_m P$$

Where, H = Enthalpy of the chemical mixture

V_m = Molar Volume

P = Internal pressure

CHAPTER 5

Results and Discussion:

The values of Ultrasonic Velocity, Density and Viscosity are measured experimentally. The variation of these parameters with mole fraction of n-Butanol is plotted. Other Acoustical parameters like Adiabatic Compressibility, Intermolecular Free Length, Acoustic Impedance, Attenuation, Relaxation Time, Effective Molecular Mass, Free Volume, Molar Volume, Available Volume, Wada's Constant, Rao's Constant, Vander wall's Constant, Internal Pressure, Gibb's Free Energy and Enthalpy are computed with the help of standard parameters and their variation is shown in the plots with respect to the mole fractions.

Table 5.1: Experimental values of Ultrasonic velocity, Density and Viscosity for ternary mixture of n-Butanol, Water and Acetic Acid at different mole fraction at temperature 289K.

Mole Fraction		Ultrasonic Velocity (U) (m/s)	Density (ρ) (kg/m^3)	Viscosity (η) (kg/ms) (10^{-3})
X_1	X_2			
0.50	0.33	1238	929.958	2.275
0.53	0.30	1363	926.586	2.025
0.57	0.26	1356	916.644	1.795
0.60	0.23	1348	900.391	1.831
0.63	0.20	1338	898.230	1.693
0.66	0.17	1340	891.746	1.879
0.70	0.13	1322	891.573	1.787
0.73	0.10	1306	876.185	2.043
0.77	0.06	1287	882.150	1.610
0.80	0.03	1278	862.461	1.590

The values of ultrasonic velocity are measured at frequency 2MHz at different mole fractions. Ultrasonic velocity, Density and viscositydecrease with increases in concentration of n-Butanol. Decrease in velocity can be observed from **Figure 5.1**. This is due to the increase in Intermolecular Free length and the Adiabatic Compressibility [15]. This behavior is different from the ideal mixture behavior and this can be attributed to the molecular association and structural changes occurring in the system [12].

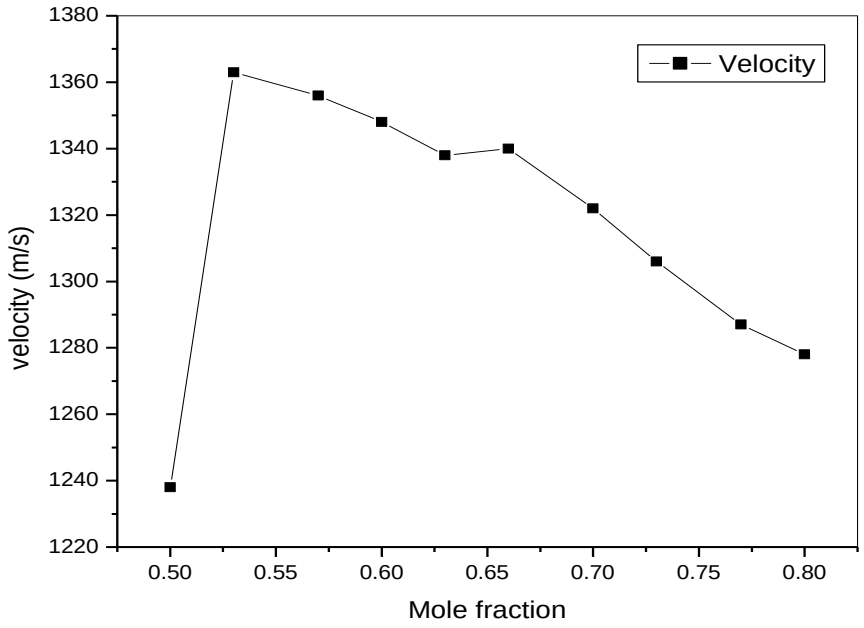

Figure 5.1: Ultrasonic velocity versus mole fraction at temperature 289 K.

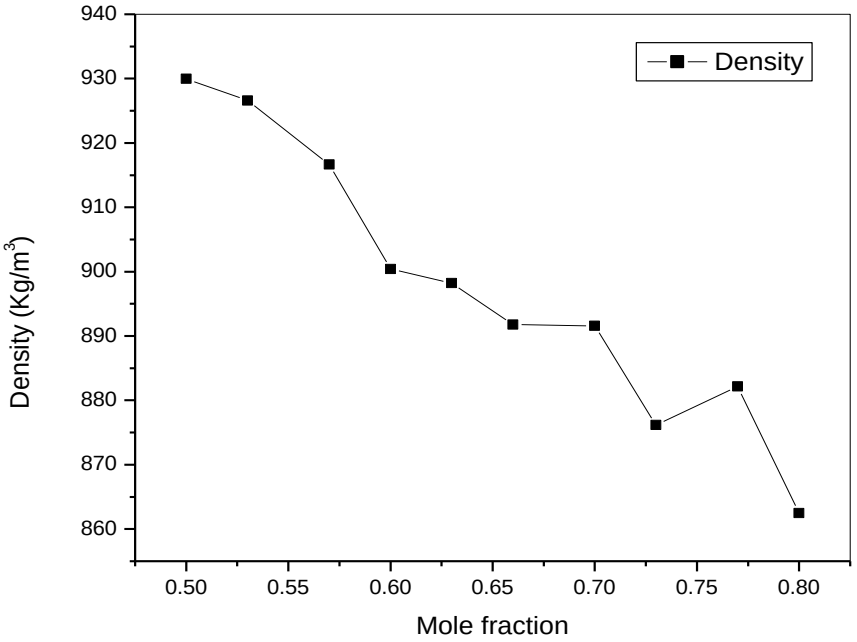

Figure 5.2: Density versus mole fraction at temperature 289 K.

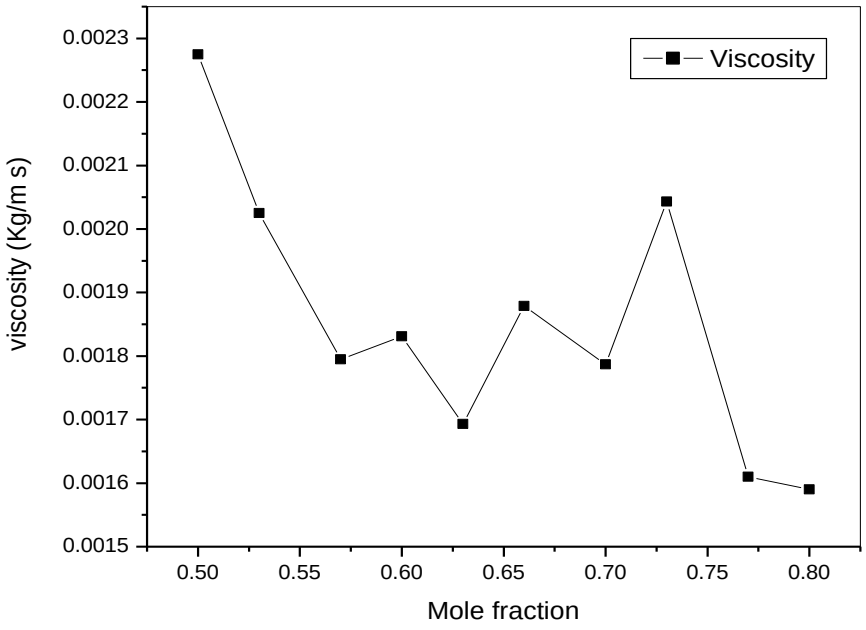

Figure 5.3: Viscosity versus mole fraction at temperature at 289 K.

Variation of viscosity and density is shown in the **Figure 5.2** and **5.3**. Density is a measure of solvent-solvent and ion-solvent interactions. Non-linear behavior is observed and shows decreases with increase in mole fraction. Decrease in density emphasizes the lesser magnitude of solute-solvent and solvent-solvent interactions. It indicates weak molecular interaction between the chemical components. The variation of density with mole fraction indicates the structural rearrangement of the molecules. Also, this increase in density with mole fraction indicates structure-breaker of the solvent [25].

From the experimentally measured standard parameters Adiabatic Compressibility, Intermolecular Free Length, Acoustic Impedance, Relaxation Time and Ultrasonic Attenuationare calculated. The values are shown in **Table 5.2**. It is clear from the table that Acoustic Compressibility and Intermolecular Free Length increases with increase in mole fraction. Both parameters are inversely proportional to the square of Ultrasonic Velocity. The trends of plots are also just opposite to that of Ultrasonic Velocity.Both of these terms decide the behavior of Ultrasonic Velocity in the

system.The increase in free length is due to the loose packing of the molecules inthe system, which interprets the presence of weak molecular interactions.

Table 5.2: Adiabatic Compressibility, Intermolecular Free Length, Acoustic Impedance, Relaxation Timeand Ultrasonic Attenuation for ternary chemical mixture at temperature 289 K.

Mole Fraction		Adiabatic Compressibility (β) (N/m^2) (10^{-10})	Intermole-cular Free Length (L_f) (A^o)	Acoustic Impedance (Z) (kg/m^2s) (10^5)	Relaxation Time (τ) (s) (10^{-12})	Ultrasonic Attenuation (α/f^2) (s^2/m) (10^{-14})
X_1	X_2					
0.50	0.33	7.018	0.5554	11.513	2.129	3.390
0.53	0.30	5.811	0.5054	12.629	1.569	2.269
0.57	0.26	5.935	0.5107	12.430	1.420	2.065
0.60	0.23	6.112	0.5183	12.137	1.492	2.083
0.63	0.20	6.219	0.5228	12.018	1.404	2.069
0.66	0.17	6.246	0.5240	11.950	1.565	2.303
0.70	0.13	6.418	0.5311	11.787	1.529	2.281
0.73	0.10	6.693	0.5424	11.443	1.823	2.752
0.77	0.06	6.845	0.5485	11.353	1.469	2.251
0.80	0.03	7.097	0.5585	11.022	1.505	2.332

From **Table 5.2** is can be observed that Acoustic Impedance decreases with increase in mole fraction. It can be defined as the ratios of Effective Sound Pressure and Effective Particle Velocity at a particular point. **Figure 5.6** illustrate the non-linear behavior of Acoustic Impedance with mole fraction. It exhibits opposite trend as compared to Adiabatic Compressibility. This is another evidence of the presence of weak molecular interactions.

Relaxation Time decreases with increase in mole fraction. From **Figure 5.7**, trend of this parameter versus mole fraction can be noted. The best results are obtained between 0.63 to 0.70 ranges of mole fraction. This behavior is due to molecular rearrangement in the mixture and the reason for this is co-operative processes.

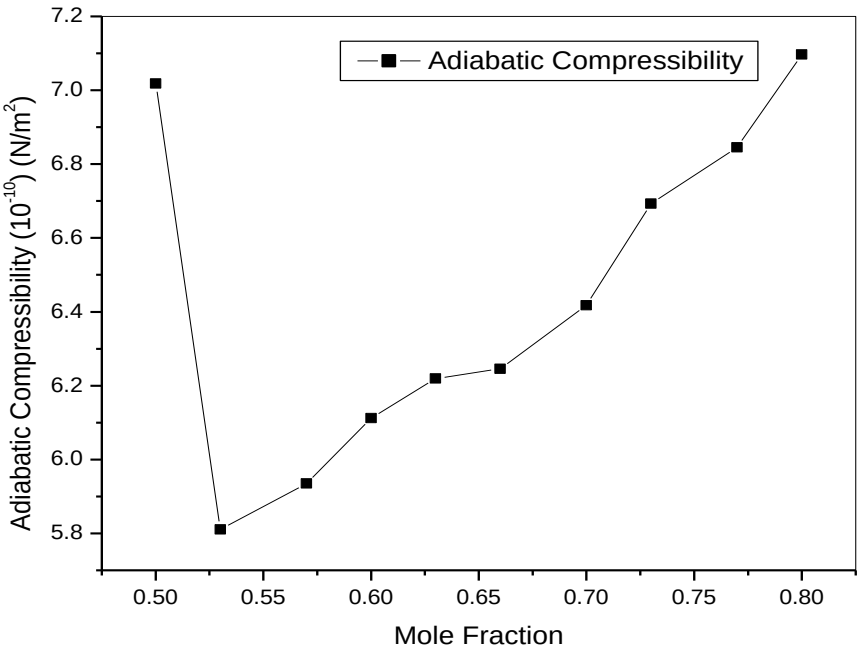

Figure 5.4: Adiabatic Compressibility versus Mole Fraction at temperature 289 K.

Figure 5.8 emphasize that Ultrasonic Attenuation shows similar behavior as Relaxation Time shows. It is a non-linear pattern. The combined effect of scattering and absorption is called **attenuation**. Ultrasonic attenuation is the decay rate of the wave as it propagates through material.

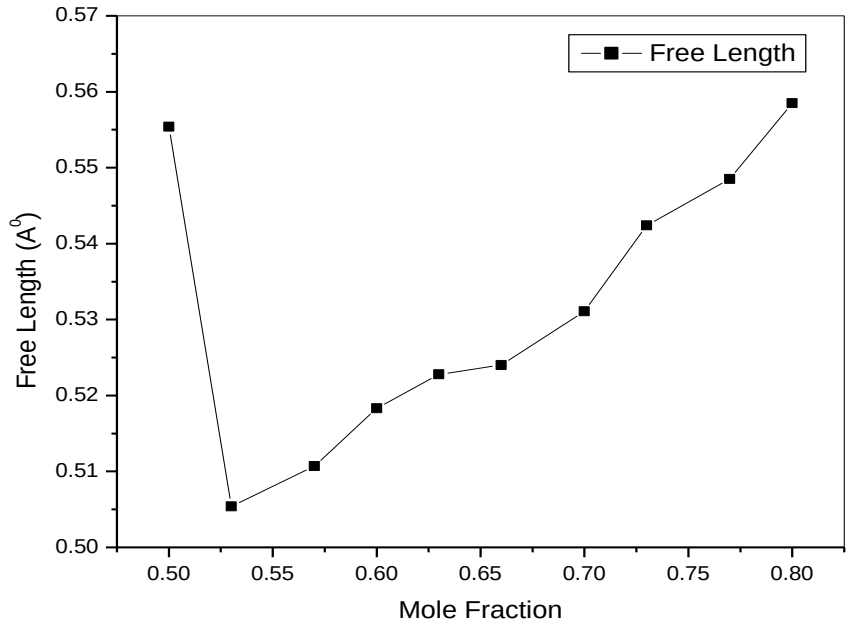

Figure 5.5: Intermolecular Free Length versus Mole Fraction at temperature 289 K.

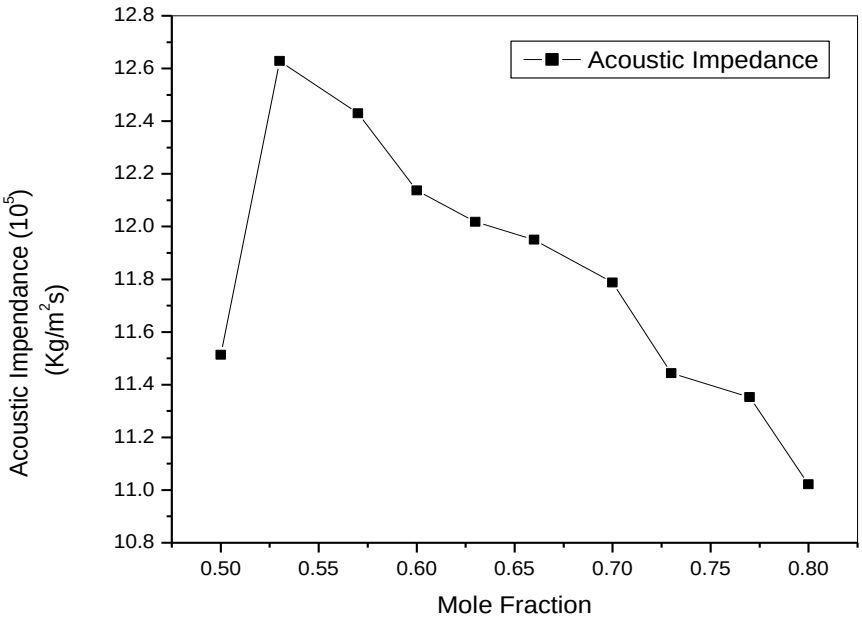

Figure 5.6: Acoustic Impedance versus Mole Fraction at temperature289 K.

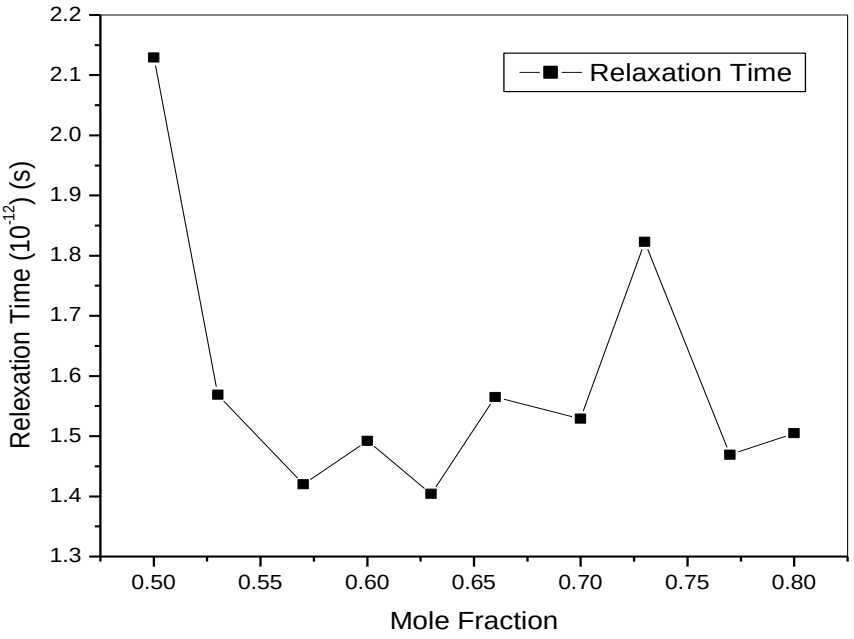

Figure 5.7: Relaxation Time versus Mole Fraction at temperature 289 K.

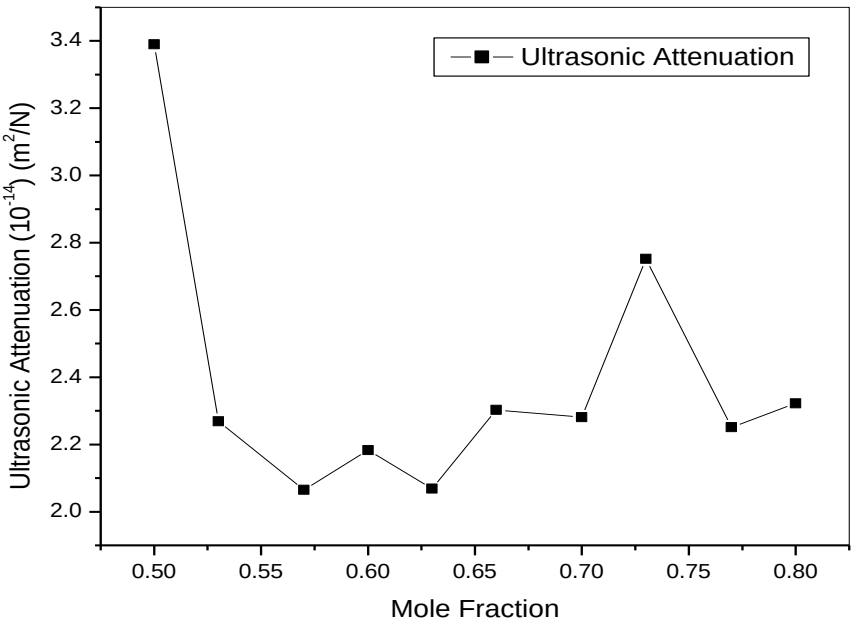

Figure 5.8: Ultrasonic Attenuation versus Mole Fraction at temperature 289 K.

Table 5.3: Effective Molecular Weight, Free Volume, Wada's Constant and Rao's Constant at temperature 289 K.

Mole Fraction		Effective Molecular Weight (M) (gm/mol)	Free Volume (V_f) (m^3/mol) (10^{-7})	Wada's Constant (W) (m^3/mol)(Pa)$^{1/7}$ (10^{-3})	Rao's Constant (R) (m^3/mol)(m/s)$^{1/3}$ (10^{-4})
X_1	X_2				
0.50	0.33	53.14	0.1756	1.160	6.136
0.53	0.30	54.80	0.2531	1.234	6.560
0.57	0.26	57.06	0.3196	1.295	6.890
0.60	0.23	58.74	0.3212	1.351	7.207
0.63	0.20	60.42	0.3727	1.390	7.412
0.66	0.17	62.10	0.3328	1.438	7.677
0.70	0.13	64.34	0.3709	1.484	7.920
0.73	0.10	66.02	0.3096	1.541	8.236
0.77	0.06	68.26	0.4552	1.577	8.417
0.80	0.03	69.94	0.4760	1.644	8.800

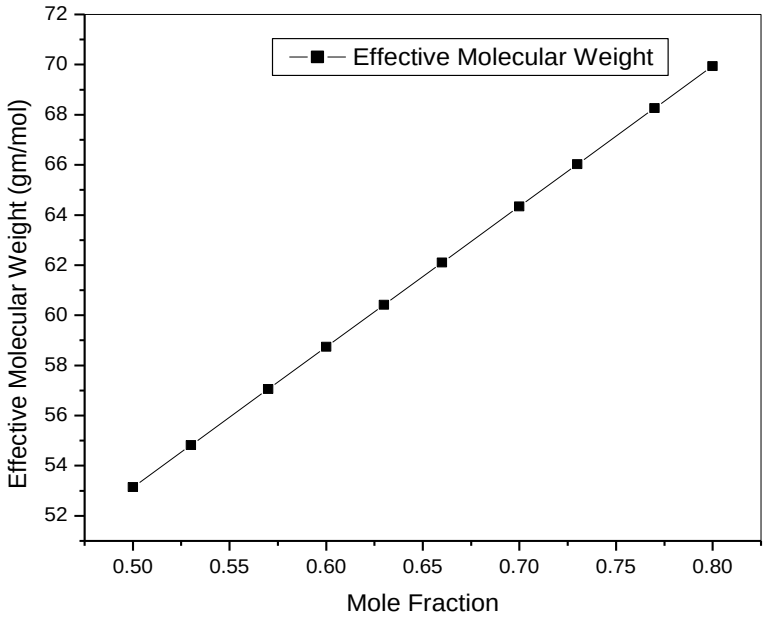

Figure 5.9: Effective Molecular Weight versus Mole Fraction at temperature 289 K.

Table 5.3 gives values of Effective Molecular Weight, Free Volume, Wada's Constant and Rao's Constant. As the mole fraction increases, effective molecular weight also increases linearly. Free Volume represents the average volume in which the centre of the molecules can move inside the cell due to the repulsion of surrounding molecules. This parameter is inversely proportional to viscosity and it can be noted from **Figure 5.10** that Free Volume increases with increase in mole fraction. The best trend can be observed between 0.63 to 0.70 ranges of mole fraction.

Figure 5.11 and **Figure 5.12** shows that Wada's Constant and Rao's Constant increase with increase in mole fraction. The trends are almost linear and this signifies the absence of complex formation in the experimental chemical system.

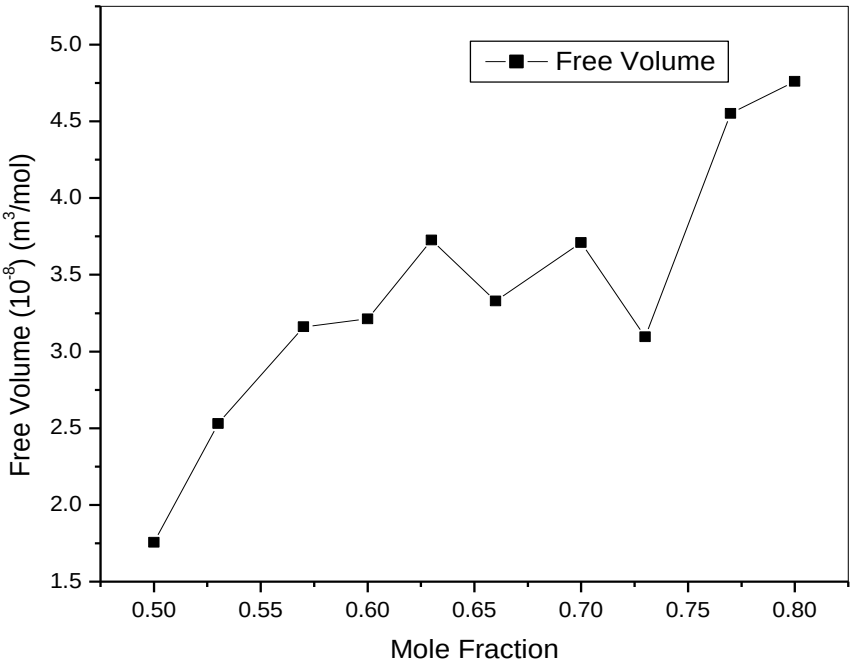

Figure 5.10: Free Volume versus Mole Fraction at temperature 289 K.

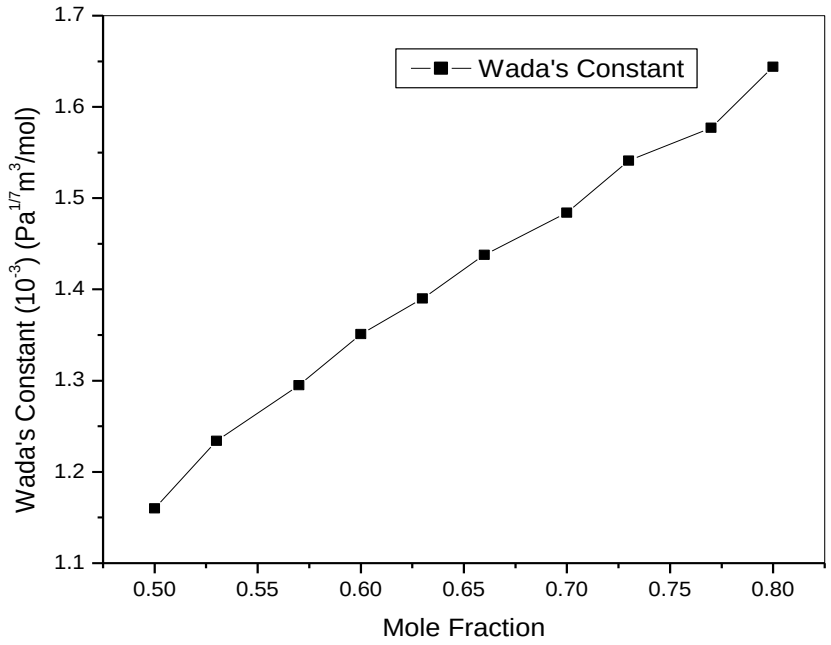

Figure 5.11: Wada's Constant versus Mole Fraction at temperature 289 K.

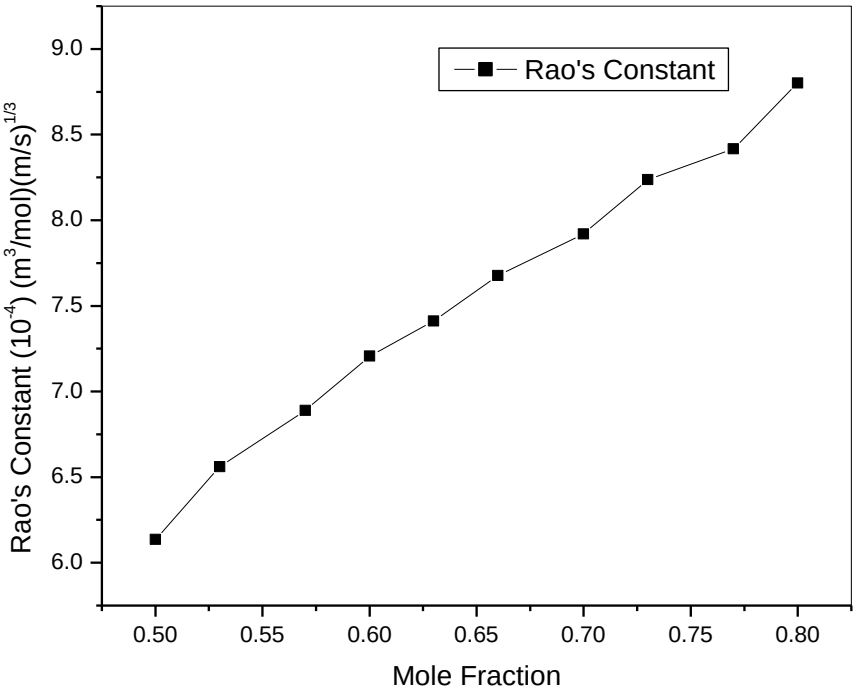

Figure 5.12: Rao's Constant versus Mole Fraction at temperature 289 K.

Table 5.4: Molar Volume, Available Volume, Vander Waal's Constant, Internal Pressure, Gibb's Free Energy and Enthalpy at temperature 289 K.

Mole Fraction		Molar Volume (V_m) (m^3/mol) (10^{-5})	Available Volume (V_a) (m^3/mol) (10^{-5})	Vander Waal's Constant (b) (m^3/mol) (10^{-5})	Internal Pressure (P) (N/m^2) (10^7)	Gibb's Free Energy (ΔG) (KJ/mol) (10^{-21})	Enthalpy (H) (J/mol)
X_1	X_2						
0.50	0.33	5.714	1.293	5.292	6.623	10.172	3784.254
0.53	0.30	5.916	0.876	5.513	5.909	8.955	3496.270
0.57	0.26	6.225	0.949	5.805	5.501	8.559	3424.509
0.60	0.23	6.524	1.027	6.087	5.480	8.755	3575.089
0.63	0.20	6.727	1.102	6.278	5.256	8.512	3575.424
0.66	0.17	6.964	1.131	6.505	5.481	8.945	3817.039
0.70	0.13	7.216	1.254	6.742	5.349	8.853	3860.245
0.73	0.10	7.535	1.385	7.040	5.664	9.554	4267.563
0.77	0.06	7.738	1.514	7.230	5.059	8.694	3915.024
0.80	0.03	8.109	1.632	7.580	4.950	8.788	4014.311

Table 5.4 shows the value of Molar Volume, Available Volume, Vander Waal's Constant, Internal Pressure, Gibb's Free Energy and Enthalpy of the chemical system. **Figure 5.13** and **Figure 5.14** shows almost linear variation of Molar Volume and Available Volume with mole fraction. As mole fraction increases, molar Volume and Available Volume also increase.Available Volume is a direct measure of the compactness and strength of bonding between the molecules of the system [13].

Figure 5.15 emphasizes the variation of Vander Waal's Constant with mole fraction. Vander Waal's Constant is also called the Co-volume and varies in Vander Waal's Equation in the similar way as Available Volume does.

Figure 5.16 unveils the relation between Internal Pressure and Mole Fraction. It shows that internal pressure decreases with increase in mole fraction. Exact trends can be noted between the mole fraction ranges from 0.63 to 0.70. This signifies that the

solutionbecomes more compressible. The reduction in internal pressure shows dissociating tendency of the molecules in the solution.

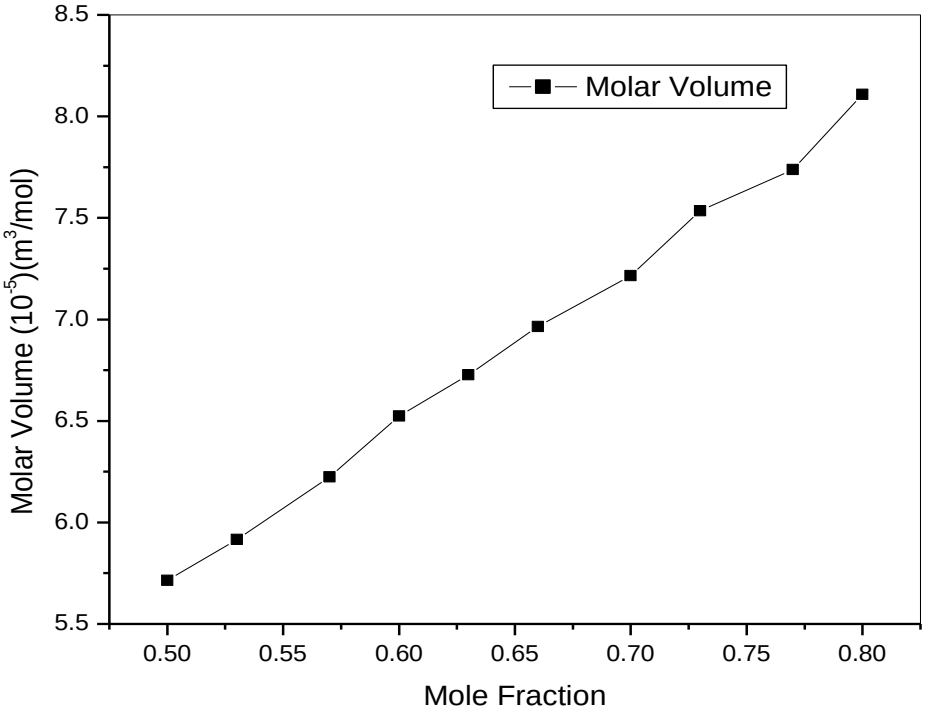

Figure 5.13: Molar Volume versus Mole Fraction at temperature 289 K.

Figure 5.17 gives non-linear relation of Gibb's Free Energy with Mole fraction. It decreases as mole fraction increases. This observation confirms the formation of intermolecular hydrogen bonding in ternary chemical mixture. Also, it gives information about the completion of the reaction. **Figure 5.18** shows the variation of Enthalpy with an increase in Mole Fraction.

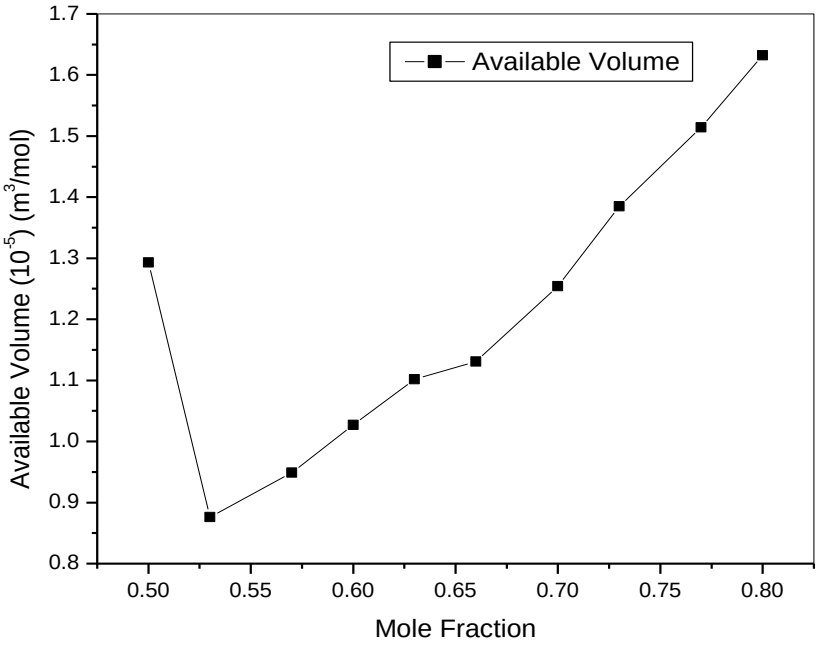

Figure 5.14: Available Volume versus Mole Fraction at temperature 289 K.

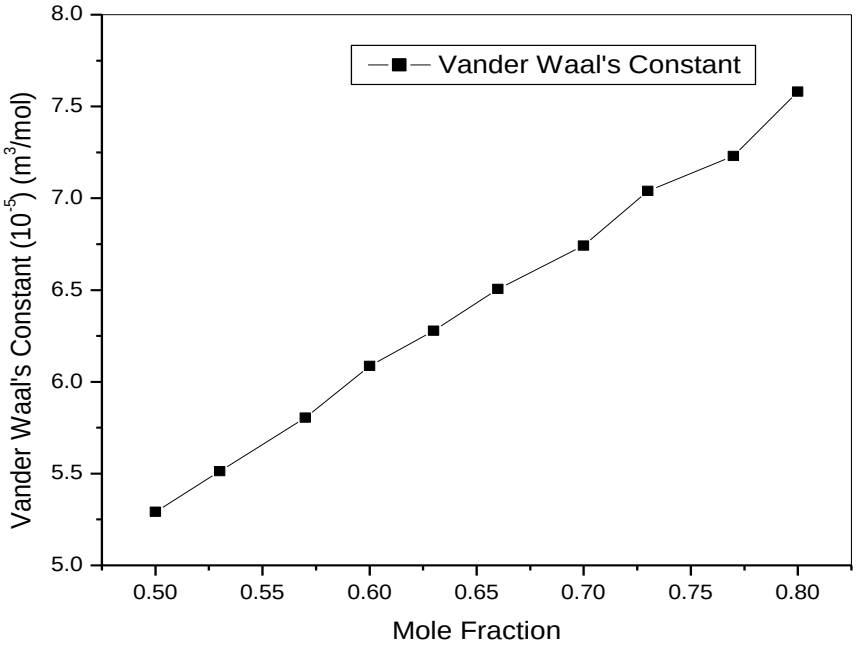

Figure 5.15: Vander Waal's Constant versus Mole Fraction at temperature 289 K.

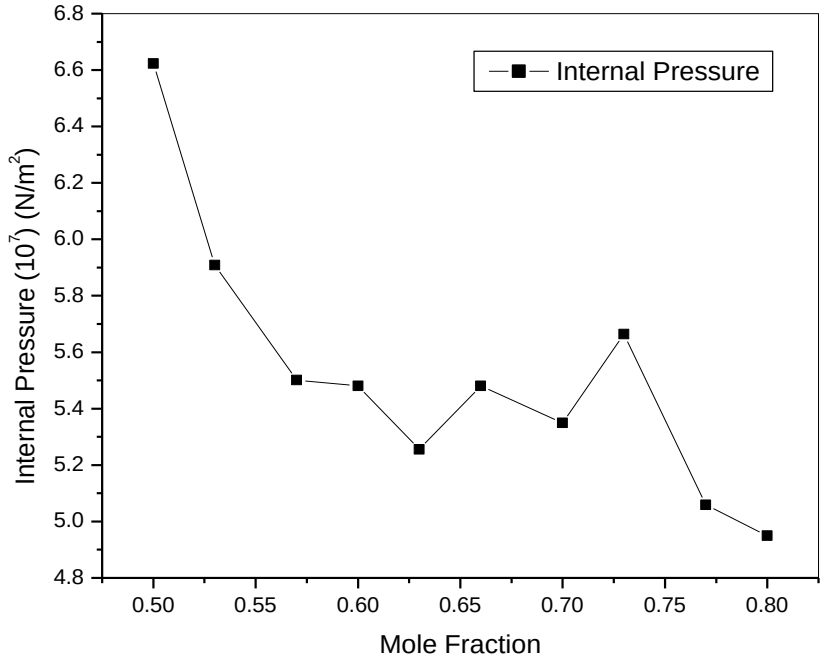

Figure 5.16: Internal Pressure versus Mole Fraction at temperature 289 K.

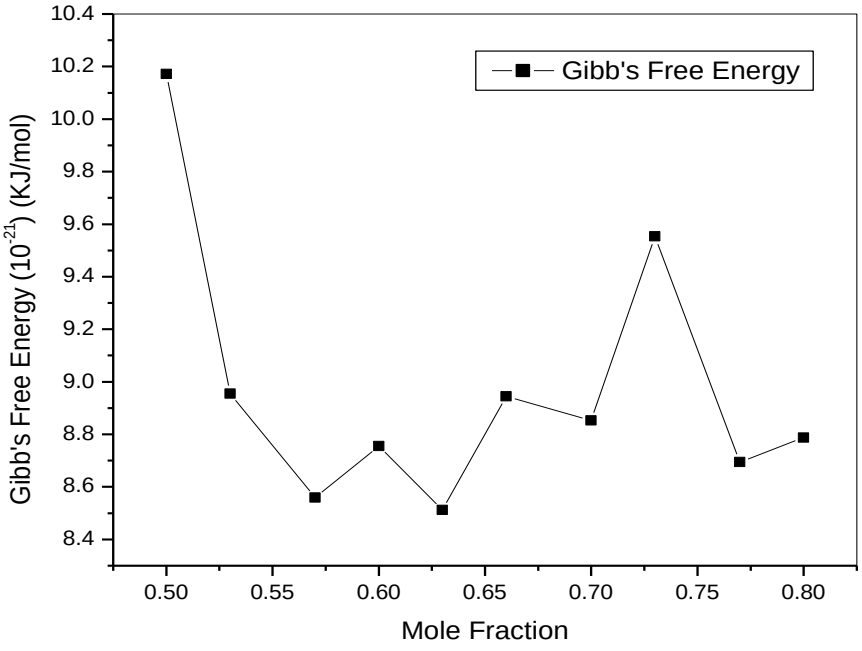

Figure 5.17: Gibb's Free Energy versus Mole Fraction at temperature 289 K.

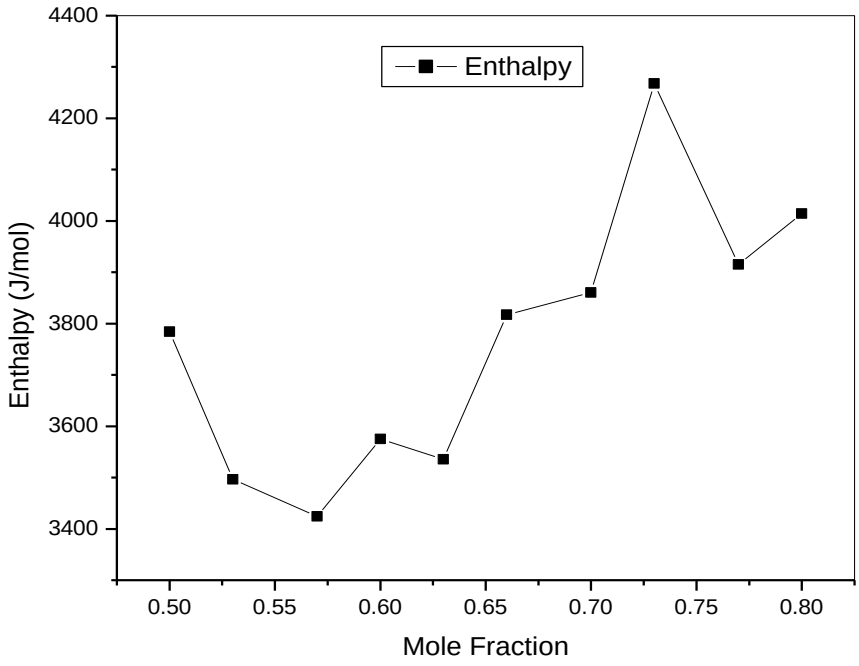

Figure 5.18: Enthalpy versus Mole Fraction at temperature 289 K.

CHAPTER 6

CONCLUSION:

In present study, the ternary chemical mixture of n-Butanol, Water and Acetic Acid forms a non ideal liquid mixture which shows considerable departure from linearity from their physical properties with respect to mole fraction. In this particular mixture presence of weak intermolecular interaction between unlike molecules is signified. Also there is solvent-solvent interaction present in the system. Complex formation in the system is completely declined. By observing the plots it can be concluded that this ternary mixture shows best results in the mole fraction range from 0.63 to 0.70. Thus between this concentration ternary mixture of n-Butanol, Acetic Acid and Water can be used as a running solvent in TLC.

CHAPTER 7

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