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Microwave Dielectric Behavior of Ayurvedic Medicines

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1. INTRODUCTION

In Material Science, characterization of materials is a significant activity. Chemical composition and structural features decides the properties of material. The properties of material also depend on the degree of molecular order.

The basics of molecular interaction are the hydrogen bonding. Hydrogen bonds occur between hydrogen containing dipoles and an electronegative element. Electro-negativity provides us a relative activity of atom in molecule to attract bonding electrons. In the present work interaction of Hydroxyl –OH group in Ethanol and Methanol at 15^0C, 25^0C ,35^0C and 45^0C is studied. In the present work interaction of Ayurvedic Medicines (Arishta group) such as Ashokarishta, Punarnvarishta and Dashmularishta from Arishta group are taken with Ethanol and Methanol.

Time Domain Spectroscopy (TDS) technique gives information in a wide frequency range from 10 MHz to 20 GHz. In the present work, reflected part of the pulse is used to obtain dielectric relaxation data. Prof. Cole developed this technique. It is very useful, economic and fast as compared to other techniques. It requires very small amount of sample and in single measurement we get permittivity and dielectric loss over wide range of frequency.

The Hewlett Packard HP 54750 sampling oscilloscope with HP 54754A TDR plug in module has been used. The TDR setup consists of step generator, sampling head, sample cell and broadband storage oscilloscope. A fast rising step pulse from generator propagates through coaxial transmission line and reaches dielectric sample placed in sample cell connected as open-ended load. It is partly transmitted and partly reflected at air dielectric interface. Both reflected as well as transmitted step from sample contains information about dielectric behavior of sample. In the present work reflected step with and without sample is recorded in the oscilloscope. This time domain data is transformed into frequency domain data using Fourier transformation. Frequency domain data is used to obtain complex reflection coefficient $\rho^*(\omega)$ over frequency range of 10 MHz to 20 GHz. Complex reflection coefficient gives permittivity and dielectric loss in selected frequency range. But normally there occurs error in this data at higher frequency due to fringing field, multiple reflections or due to quarter wave resonance in case of high lossy liquids. The complex reflection data is called 'RAW' data. An error in 'RAW' data is corrected by bilinear calibration process.

The corrected data is called 'COR' data. The dielectric parameters of the Ayurvedic Medicines are obtained by fitting 'COR' data to Harviliak Negami equation-

$$\varepsilon^*(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\left[1 + (j\omega\tau)^{(1-\alpha)}\right]^\beta} \quad (1)$$

If we put α =0, β =1 then equation represents simple Debye model. A Least Square Fit method is used to obtain dielectric parameters.

The dielectric relaxation time (τ) of biological material is related to nature of intermolecular bonding, size of molecule, mobility of molecules in solutions, molecular volume, viscosity and temperatures similarly, the permittivity(ε) is related to square of molecular dipole moment, and the value of permittivity(ε) is related to size of molecule in solution and temperatures. Thus information at molecular level can be gained from a study of dielectric behavior. To understand structural change in system Excess Permittivity (ε^E), Excess inverse relaxation time $(1/\tau)^E$ and Bruggeman factor (f_B) are obtained. Thermodynamic parameters i.e. activation energy in KJ/mole, change in enthalpy (ΔH) and change in entropy (ΔS) are calculated using Eyring's equation to understand molecular dynamic of the system.

2. AYURVEDA

Ayurveda[1] is a holistic healing science, which comprises of two words, Ayu and Veda. Ayu means life and Veda means knowledge or science. So the literal meaning of the word Ayurveda is the science of life. One of the basic principles of Ayurveda, the recognition of the inner reflecting the outer.

By using ayurvedic and herbal medicines you ensure physical and mental health without side effects. The natural ingredients of herbs help bring "arogya" to human body and mind. ("Arogya" means free from diseases). The chemicals used in preparing allopathy medicines have impact on mind as well. One should have allopathy medicine only when it is very necessary. According to the original texts, the goal of Ayurveda is prevention as well as promotion of the body's own capacity for maintenance and balance. Ayurvedic treatment is non-invasive and non-toxic, so it can be used safely as an alternative therapy or alongside conventional therapies. Ayurvedic physicians claim that their methods can also help stress-related, metabolic, and chronic conditions. Ayurveda has been used to treat a acne, allergies, asthma, anxiety, arthritis, chronic fatigue syndrome, colds, colitis, constipation, depression, diabetes, flu, heart disease, hypertension, immune problems, inflammation, insomnia, nervous disorders, obesity, skin problems, and ulcers.

3. DIELECTRIC STUDY IN MEDICINE

The recent application of dielectric studies on biological molecules has shown the possibility of treating malignant diseases by studying the responses of diseased cells of the body to the radiation used and then localizing the heat in the area of interest. The energy necessary for hyperthermia of a 20 mm diameter tumor by 5^{o}C in one minute, if energy is efficiently transmitted to the tumor. Neoplastic tissue has a higher dielectric loss factor than healthy

tissue because of its higher water content. For example, skin carcinoma contains 81.6% as compared with 60.9% in normal epidermis; hepatoma contains 81.9% as compared with 71.4% in liver. These differences are sufficient to enable us to discriminate between different types of carcinoma by diagnostic radio – frequency imaging.

The information such as molecular flexibility or rigidity, shape and size etc. obtained using dielectric relaxation as the probe which serves the basis for determining its carcinogenic or anti-carcinogenic action, is of vital importance for investigation of cancer.

The values of dielectric constant and dielectric relaxation time for mouth cancer patient's saliva are ($\varepsilon_0 = 81.68$, $\tau = 13.29$ ps) larger than normal person's saliva ($\varepsilon_0 = 76.57$, $\tau = 11.38$ ps).Now a days, popularity of Ayurvedic Medicines becomes worldwide. Ayurvedic Medicines used in Gyneac problems in human body are considered in the present work.

Dielectric spectroscopy is a branch of spectroscopy where one gets information about structural changes and molecular interactions through dielectric relaxation data. In the present work interaction of Ayurvedic Medicine such as Ashokarishta, Punarnvarishta, and Dashmulrishta from Arishta group are taken with Ethanol and Methanol

4. DIELECTIRC RELAXATION SPECTROSCOPY

Dielectric Relaxation Spectroscopy (DRS) probes the interaction of a macroscopic sample with a time-dependent electric field[2]. The resulting polarization either expressed by the frequency-dependent complex permittivity and conductivity or as an impedance spectrum, characterizes amplitude and timescale (via the relaxation time) of the charge-density fluctuations within the sample. Such fluctuations generally arise from the reorientation of the permanent dipole moments of individual molecules or from the rotation of dipolar movements in flexible molecules, like polymers. Other possible mechanisms include the transport of ions or the appearance of interfacial charges in heterogeneous systems. The timescale of these fluctuations depends on the sample and on the relevant relaxation mechanism. Relaxation times range from several picoseconds in low-viscosity liquids to hours in glasses, probably marking DRS as the technique with the most extensive coverage of dynamical processes. The corresponding measurement frequencies range from 10^{-4} Hz to 10^{12} Hz, which requires a series instruments for complete coverage. However, it is generally sufficient to concentrate on a smaller frequency range adapted to the sample properties. In contrast to conventional spectroscopic methods, like NMR or vibration spectroscopy, DRS is especially sensitive to intermolecular interactions. DRS is able to monitor cooperative processes and thus provides a link between molecular spectroscopy, which monitors the properties of the individual constituents, and techniques characterizing the bulk properties of the sample, especially the viscoelastic and archeological behavior. The decomposition of the dielectric spectrum into its individual relaxation processes informs on the relative amplitudes and characteristic times of the underlying molecular motions. Dielectric relaxation studies on binary mixture are important for understanding the hydrogen bonding and intermolecular interaction in the mixture. The dielectric relaxation study of solute – solvent mixture of microwave frequency gives information about molecular interaction in the system, formation of monomers and multimers. Dielectric Spectroscopy is being successfully used to determine the time of relaxation of electrolytes in solution.

DRS is widely applied in the characterization of ion-conducting solids, polymers and mesophases. But it is also of large potential interest for the investigations of liquid and colloidal systems. Additionally, the effects studied by DRS are of increasing importance for technical applications like dielectric heating or remote sensing.

5. DIELECTRIC POLARIZATION

When a dielectric is placed between charged plates, the polarization of the medium produces an electric field opposing the field of the charges on the plate. The dielectric constant k is defined to reflect the amount of reduction of effective electric field . The permittivity is a characteristic of space, and the relative permittivity or "dielectric constant" is a way to characterize the reduction in effective field. Because of the polarization[1] of the dielectric. The capacitance of the parallel plate arrangement is increased by factor k.

According to spatial arrangement of charges in a molecule the molecules are classified as polar and non-polar[6]. A polar molecule has permanent dipole moment. The dipole moment depends on the size and symmetry of the molecule. Although the total number of positive and negative charges is equal to the distribution of two kinds of charges is different. Non-polar dielectric consists of molecules with positive and negative charges such that their effective center of charge distribution coincides. Thus dipole moment of non-polar dielectric material is zero in absence of electric field.

If a distance ‘d’ separates the charge +q and -q, it forms a dipole moment given by - qd. In a molecule 'q' is of the order of electronic charge, 10^{-10} e.s.u., while the 'd' is of the order of 10^{-8} e.s.u. Therefore unit of dipole moment is 10^{-18} e.s.u., and is called a 'Debye', abbreviated as 'D'. In the case of non-polar molecules, the centers of positive and negative charges coincide with the centers of symmetry of the molecule, therefore they have zero dipole moment. e.g. Benzene, Methane. Polar molecules always have a permanent dipole moment, even in the absence of an external electric field.

When the electric field is applied to dielectric, the molecular charges get displaced. The total charge passing through unit area within the dielectric, perpendicular to the direction of applied field is called polarization. The polarizations are of three types, Electronic Polarization (P_e), Atomic Polarization (P_a), Orientation Polarization (P_o).

Orientation polarization is property of the polar molecules. It is due to rotation of permanent dipoles of dielectric medium. The molecular dipoles orient in the direction of the applied field. It is function of molecule size, viscosity, temperature, and frequency of applied field. Orientation polarization takes a time of the order of 10^{-12} to 10^{-10} sec., corresponding to period of microwave frequency region. The total polarization is,

$$P_t = P_e + P_a + P_o \tag{2}$$

Thus the polar materials have greater permittivity than the non-polar, because of additional polarization due to orientation The insulator whose behavior gets modified in the electric field are called as dielectric. Dielectric materials are bad conductors of electricity. When these materials are placed in dielectric field, displacement of positive and negative charges in molecule takes place. When the change in the behavior of dielectric is independent of the direction of the applied field, the dielectric is called Isotropic. On the other hand if the change in behavior of dielectric depends on the direction of applied field the dielectric is called anisotropic. The positive and negative charge distribution separated by some distance

can be treated as dipole. Applied electric field forces this molecular dipole to align in the direction of field. This alignment of molecular dipoles in the direction of the field is called polarization dielectric constant . It is a measure of ability of material to get polarized in the direction of applied electric fields. Hence dielectric material store applied electrical energy in the form of polarization.

6. STATIC AND DYNAMIC PERMITTIVITY

The theories of dielectric relaxation can be broadly divided into two parts as theories of static permittivity and theories of dynamic permittivity. The polar dielectric materials having a permanent dipole moment, when placed in steady electric field so that all types of polarization can maintain equilibrium with it, the permittivity of material under these conditions is called as static permittivity (ε_o), when dielectric material is placed in electric field varying with frequency, then permittivity of material changes with change in frequency of applied field. This is so because with increasing frequency molecular dipoles cannot orient faster to come up with applied field. Thus permittivity of material falls off with frequency of applied field, the frequency dependent permittivity of material is called as dynamic permittivity The different theories of static and dynamic permittivity like Clausius Mossotti Equation, Debye Theory of Static Permittivity, Onsager Theory of Static Permittivity, Frohlich's theory are used as well as, the Debye Model ,the Cole-Cole Model,the Davidson-Cole Relaxation Model,the Havriliak-Negami Model are used.

Dielectric relaxation occurs when; the externally applied alternating field polarizes a dielectric material. The decay in polarization is observed on removal of the field. The decay in polarization occurs due to orientation of electric dipoles in an electric field. This depends on the internal structure of a molecule and on molecular arrangement. The orientation polarization decays exponentially with time; the characteristic time of this exponential decay is called relaxation time. It is defined as the time in which this polarization reduces to $(1/\tau)^{th}$ times the original value. Dielectric relaxation is the cause of anomalous dispersion in which permittivity decreases with increasing frequency.

Under the influence of an ac electric field, the polar molecules of a material orient themselves and attain an equilibrium distribution in molecular orientation. When the polar molecules are of large size or frequency of ac field is very high or the viscosity of the medium is very large, the orientation of molecules is not fast enough for the attainment of equilibrium with the applied field. The polarization then acquires a component out of phase with the field and the displacement current acquires a conductance component in phase with field, resulting in thermal dissipation of energy. The permittivity thus acquires a complex characteristic.

In such cases it is used to relate the displacement $\vec{D} = \varepsilon^* \vec{E}$. The complex permittivity ε^* can be written as $\varepsilon' - i\varepsilon''$, where ε' is real part proportional to stored energy and ε'' is imaginary part and it is dielectric loss.

7. TDR TECHNIQUE

Time Domain Spectroscopy has become a widespread method of investigation for variety of substances. Dielectric study provides information about charge distribution in a molecular

system. The recently developed Time Domain Reflectometry has proved to be very effective and efficient for determination of dielectric constant and loss in the frequency range of 10 MHz to 10 GHz. Time Domain Reflectometry (TDR) method is the most suitable method for determination of frequency dependent dielectric parameters of material. This method was first introduced by Hugo - Fellner –Feldegg[5] (et al in 1969) and developed by many workers in field of dielectric spectrometry.. TDR technique is being adopted to measure static conductivity of electrolytic solution.

In TDR method, a fast rising step pulse is allowed to incident on sample under investigation. The reflected pulse from sample contains the information regarding dielectric behavior of sample. The Fourier transformation of step pulse gives us frequency components contained in step pulse. Thus incident step pulse is treated as mixture of waves with different frequencies. The lower limit of frequency spectrum contained in step pulse depends on time window used, while upper limit depends on rise time of pulse. Frequency dependent permittivity parameters of sample can be determined from its response to incident step pulse.

In order to obtain frequency dependent dielectric parameters one needs a step generator, a sampling oscilloscope, a sample holder and mathematical expression with computer software. The step generator must be capable of generating step pulse with rise time adequate enough to give the highest frequency components of interest with considerable magnitude. The broadband oscilloscope is required to handle broad frequency spectrum contained in step pulse with sufficient accuracy. A transmission line is needed to carry signal from step generator to sample holder. The transmission line as well as sample holder must be capable of holding high frequency signals.

For faithful transmission characteristics impedance of components must be matched. Any impedance mismatching in this signal path carries multiple reflection, which can disturb signal of our interest. Practically multiple reflection of signals, when it passes from one component to other cannot be avoided totally, but can be minimized to acceptable level by making same precautions. The time domain data is converted into frequency domain data using Fourier transformation.

Experimental setup consists of sampling oscilloscope HP 54750A, TDR module HP 54754A, a transmission line and sample cell. The HP 54750A-sampling oscilloscope is very precise instrument for digital data acquisition of instantaneous signals. The working of instrument depends on front panel keys as well as menus invoked after pressing any front panel key. The menus of function are displayed along the right side of display screen. These menus are called soft key menus. Soft key menus list functions other than those accessed directly by the front panel keys. To activate a function on soft key menu can be accessed by pressing unlabeled key immediately next to the annotation on the screen. The unlabeled keys next to the annotation on display are called soft keys. Front panel of the instrument includes a display area and several functional areas, which includes control, storage, autoscale, entry devices, setup, and system. Control section includes three keys clear display, run and stop signal. These keys are used to clear screen, start data acquisition and stop data acquisition respectively. Storage section includes four keys disk, waveform, setup and print. Disk key is used to access information from 1.44 MB Floppy Disk Drive. We can store the waveforms on disk or load waveform from disk. Waveform key is used to store current waveform in memory of oscilloscope. Four waveforms can be stored at a time in oscilloscope memory. Setup is used for setting waveform. Print key is used to print current waveform or

waveform in memory. Autoscale section contains only single key Autoscale. This Autoscale key causes the instrument to quickly analyze the signal. Then, it sets up vertical, horizontal and triggers to best display that signal. Entry devices are the keypad, the arrow keys and the knob. Entry devices can change the numeric settings of some soft-keys, such as trigger level, or to select an item from the list of choices. The setup section includes seven keys. Time base, Trigger, Acquisition, Display, marker, Define meas. and Math. With time base key we can change horizontal position of waveform and also the time window. Trigger can be used to change trigger level of signal. Acquisition key is used to set number of data acquisition points and also number of times the averaging is done. Marker key can be used for setting markers on waveform during measurement of specific parameters. One can also put meas. (measurement marker lines) during measurement. Math function key is used to perform few mathematical operations such as addition and subtraction of two waveforms or even Fourier transform of waveform.

The HP 54754A TDR plug in module is capable of performing both, single ended TDR measurements as well as differential TDR measurements. These measurements include characterizing micro strip lines, PC board traces and coaxial cables. The plug in module takes up two, out of four mainframe slots. In single ended TDR measurement, a positive going step (a fast rising step voltage pulse of 200 mv with 39 ps rise time) is launched on one of the channels while the other channel is terminated using short. In differential TDR measurement, a positive going step is launched on channel 1 and an effective negative going is launched on channel 2. The response controls are provided which shows the single ended or differential mode response of a TDR system. A fast rising step voltage pulse of about 200 mV amplitude and 43.8486 ns rise time with repetition frequency of 12.4 GHz is generated and is propagated through a coaxial transmission line. The sample is placed at the end of the coaxial transmission line in a standard Military application (SMA) coaxial cell. The SMA cell used for this work had 3.5 mm outer diameter and 1.52 mm effective pin length. The step pulse generated by tunnel diode and the pulse which is reflected from the sample cell were sampled by a sampling oscilloscope in the time window of 1.3 ns. The reflected pulse without sample $R_1(t)$ and with sample R_x (t) averaged 64 times and digitized with 1024 points in oscilloscope memory and transferred to PC through a 1.44 floppy diskette drive.

The temperature controller system with water bath and a thermostat has been used to maintain the constant temperature within the accuracy limit of ±1 ^{0}C. The sample cell is surrounded by a heat-insulating container through which the water of constant temperature using temperature controller system is circulated. The temperature at the cell is checked using the thermometer. The combination of Ayurvedic Medicines with Ethanol and Methanol are taken at different temperatures such as 15^{0}C, 25^{0}C, 35^{0}C and 45^{0}C. No work has been reported on this systems using Time Domain Reflectometry (TDR) technique. All the above systems are studied by preparing 11 concentrations by volume fraction 'X' of solutions such as 0%, 10%, 20%,…….100% with the two pure liquids. Temperature dependent variations in dielectric parameters and thermodynamic parameters for four different temperatures are reported.

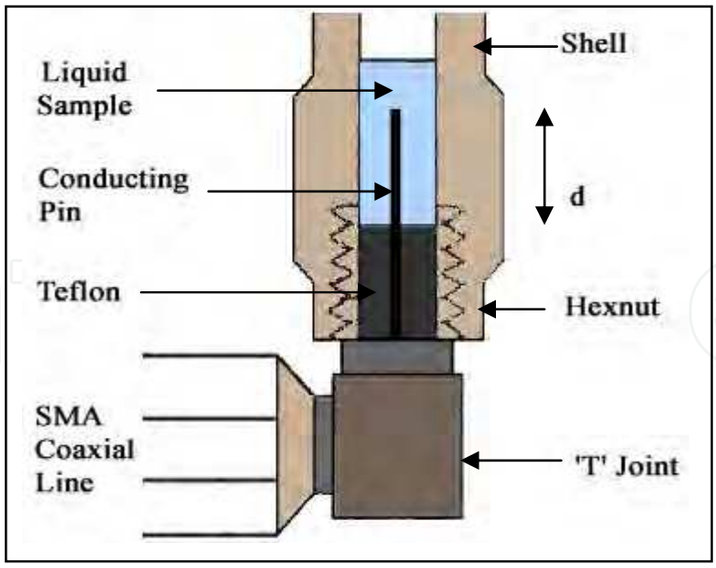

Fig.1. Geometrical construction of SMA cell

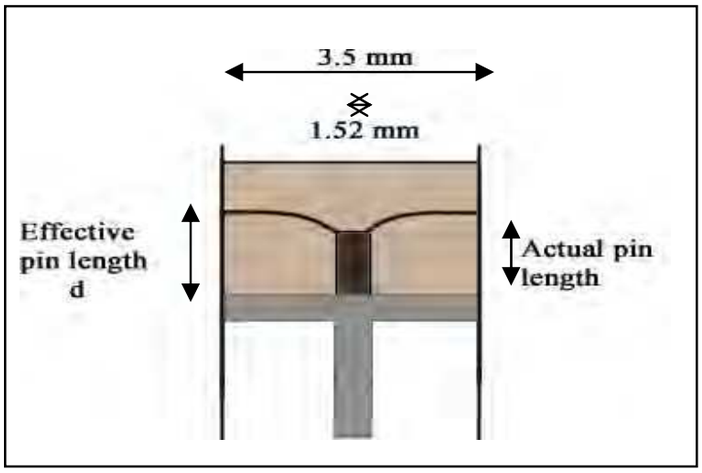

Fig. 2. Fringing field and SMA cell dimension

8. DIELECTRIC RELAXATION BEHAVIOUR OF AYURVEDIC MEDICINES

The dielectric relaxation study at microwave frequency gives information about solute – solvent interaction and liquid structure of mixture. By using Time Domain Reflectometry technique different types of liquids such as pharmacologically important drugs, n nitrites, glucose water mixtures, Binary mixtures, Biological samples (Methanol – algae), amides, Alcohols, electrolytes, liquid crystals were studied. Ayurvedic medicines from 'Arishta' group are selected to observe velatine structural changes as well as changes in interaction of these molecules with ethanol and methanol. Static permittivity, relaxation time, Bruggeman factor, Activation energy, enthalpy, entropy are reported for various systems and for different temperatures. The preparation of Ayurvedic medicine is always complex in nature, as it contains alcohol, sugar, variety of medicinal herbs, their leaves, flowers, fruits, peels,

roots, sap (gum), resins. It also contains shells, conches cowries coral and pearls found in the sea and metals like gold, silver, lead, mercury, copper or iron..

'Arishta' is prepared with the help of extract of medicinal material or juice and it is mixed with jaggery, sugar, honey, or other sweeteners. According to Ayurvedic science it is fermented, brewed for a period for 2-3 months. The process (Kinwa) of fermentation occurs at certain temperature near about 30°C to 35°C After formation of 'Arishta' percentage of alcohol is there e.g. Ashokarishta contains 7.4% of alcohol, Dashmularishta contains 8.8% of alcohol, Punarnarishta contain 6.4% of alcohol etc. The Sanskrit name of medicinal plants and metals has been used to indicate the standard names of 'Arishta' group. Eg. Ashokarishta contain 'Ashoka' as medicinal plant as well as another 14 different plants are used in a minor portion.

Dashmularishta contain 10 different roots hence the name Dashmularishta. Basically Dashmularishta is used to increase the immune system of human being. Punarnvarishta contain 'Punarna' as a medicinal plant. It is used to improve the working of heart, liver, pancreas, kidney etc. These Ayurvedic Medicines also consist of water, carbohydrates, protein, fats, alkaloid and alcohol molecules. The functional groups commonly present in these molecules are hydroxyl (-OH), aldehyde (-CHO), carbonyl (>C=O), Caboxylic (-COOH), amine (-NH-), methane (C-H) and cyanide (C-N).

Hydrogen bonding is the basics of all molecular interactions. The distinguishing feature of hydrogen bonding is the involvement of a specific H atom of a proton donar group with a localized site of high electron density in the same or another molecule. Another important feature of hydrogen bonding and of other weak attractive interactions in solution is that, at ordinary temperature, only a fraction of the molecules are generally associated. At equilibrium while a certain number of new complexes are continually formed, an equal number of complexes are continually broken due to the kinetic energy of motion of the interacting molecules. Basically hydrogen bonding occurs between a proton donar and proton accepter group. The hydroxyl (-OH), carboxyl (-COOH), amine (-NH-) and cyanide (C-N) are proton donar as well as proton accepter group. Hydrogen bonds occur between hydrogen containing dipoles and an electromagnetic element. The carboxyl group and oxygen atom have more electronegativity. Electronegativity provides us a relative ability of atom or functional group in molecule to attract bonding electrons.

The enthalpy of hydrogen bonds generally falls in the range of 1 to 10 Kcal/mole. Oxygen is a good proton accepter whether it is attached to phosphorous, to sulphur, to carbon or to nitrogen. The anions of electronegative atom from strong hydrogen bonds. Time Domain Reflectometry (TDR) is an effective approach to understand molecular interactions in liquid. Time Domain Reflectometry in reflection made is used as technique. It is very interesting to correlate dielectric parameters to molecular dynamics in aqueous solutions, hydrogen bonding change in size of molecular entities as well as their speed of rotation, in presence of different types of solutes was carried out by many research groups in the field of dielectric spectroscopy.

9. DATA ANALYSIS

The time dependent data were processed to obtain complex reflection coefficient $\rho^*(\omega)$ over the frequency range from 10 MHz to 20 GHz. Using Fourier Transformation as,

$$\rho^*(\omega) = (c/j\omega d)^* [p(\omega)/ q(\omega)] \quad (3)$$

Where p(ω) and q(ω) are Fourier transforms of $[R_1(t)- R_x(t)]$ and $[R_1(t) + R_x(t)]$ respectively, c is velocity of light, ω is angular frequency, $j = \sqrt{-1}$, d is effective pin length and the complex permittivity spectra ε*(ω) were obtained from reflection coefficient ρ*(ω) spectra by applying a bilinear calibration method[8]. The example of ρ*(ω) and ε*(ω) spectra are shown in Fig. 3 and 4 respectively.

The experimental value of ε*are fitted with the Debye equation,

$$\varepsilon^* = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)}{(1 + j\,\omega\,\tau)} \quad (4)$$

With ε_o and τ as fitting parameters. In Eq. 4, ε_o is the static permittivity, τ is the relaxation time and ε_∞ is the permittivity at high frequency. The value of ε_∞ is taken to be 3.0 for all the system studied, as for the frequency range considered here, the ε* is not sensitive with respect to ε_∞. A non-linear Least-Squares fit method used to determine the values of dielectric parameters.

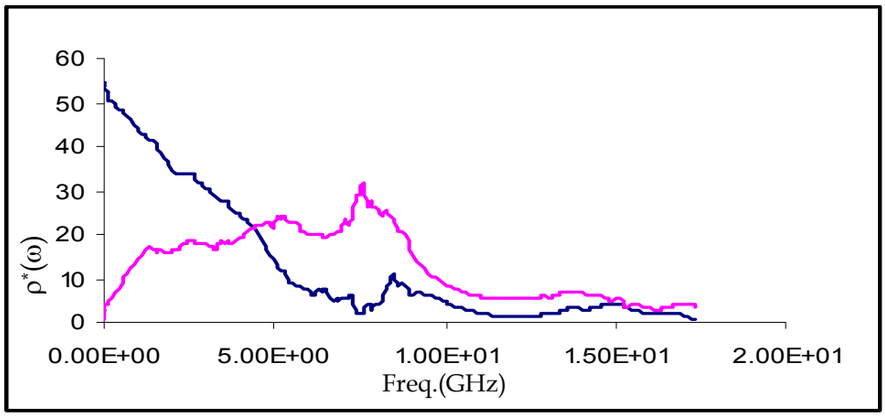

Fig. 3 ρ*(ω) spectra for 80% Ashokarishta + 20% Ethanol at 15°C

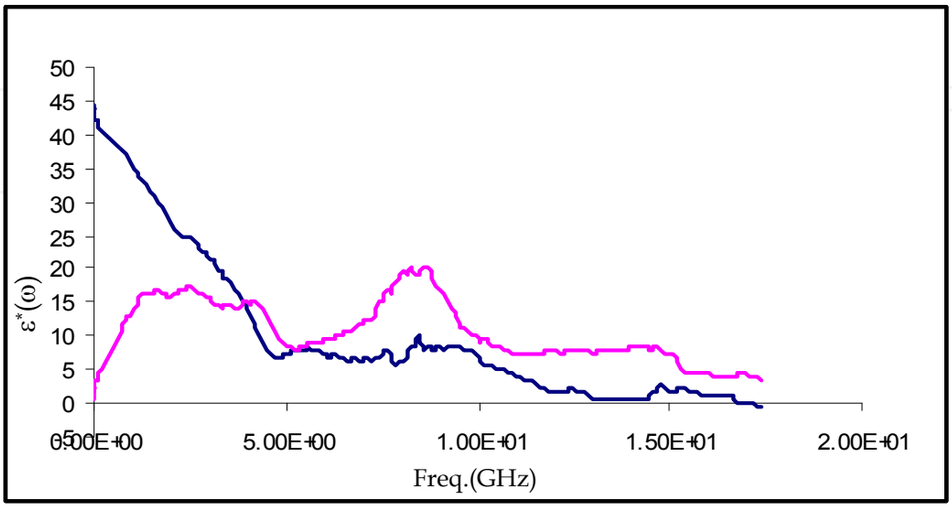

Fig. 4. ε*(ω) spectra for 80% Ashokarishta + 20% Ethanol at 15°C

10. AYURVEDIC MEDICINE 'ASHOKARISHTA'AND ETHANOL

The Frequency dependent complex permittivity spectra $\varepsilon^*(\omega)=\varepsilon'(\omega)-j\varepsilon''(\omega)$, in frequency range of 10 MHz to 20 GHz, for mixture of Ashokarishta and Ethanol is presented in this section.

Ashokarishta (Shri. Baidyanath pharmaceutical ltd., Kolkata) and Ethanol (Changshu yang Yuan Chemical, China) were obtained commercially and used without further purification. The solutions were prepared at different volume percentage of Ashokarishta in Ethanol in steps of 10% vol., within 0.01% error limit. The experiment is performed for 11 concentrations, at 15^0C, 25^0C, 35^0C and 45^0C temperatures. The relative change in dielectric parameters with increasing concentrations of solute and temperature are presented in this section. The bilinear calibration method is used to obtain complex permittivity $\varepsilon^*(\omega)$ from complex reflextion coefficient $\rho^*(\omega)$. Reflection coefficient spectra $\rho^*(\omega)$ for pure Ethanol is used to correct $\rho^*(\omega)$ of entire concentrations range at each temperature. The corrected data for each mixture is fitted to Debye equation to obtain static permittivity (ε_o) and relaxation time (τ). As the frequency range of dielectric investigation in present work is from 10 MHz to 20 GHz, the value of (ε_o) obtained is just fitting parameter. This value does not correspond to real value of permittivity which one gets after completion of dispersion processes related to vibration and electronic motions in liquid. It is found reasonably satisfactory procedure to keep value (ε_∞) fix. In the present work value of (ε_∞) is kept fix to 3.00. The corrected spectra $\varepsilon^*(\omega)=\varepsilon'(\omega)-j\varepsilon''(\omega)$ for mixtures of Ashokarishta - Ethanol system for different concentrations at 35^0C is shown in fig. 5. Gradual decrease in permittivity and dielectric loss with increasing volume fraction of Ethanol can be observed from this fig. The fall in permittivity starts at low frequency for Ashokarishta rich region but this point shifts to higher frequency in Ethanol rich region. This indicates small change in alignment of molecular dipoles with addition of Ethanol in Ashokarishta. The Cole- Cole plot for Ashokarishta and Ethanol system at 35^0C is shown in fig. 6. The Cole Cole plot follows the Debye semicircle, which indicates that relaxation process in Ashokarishta-Ethanol system can be explained with single relaxation time.

10.1 Static Permittivity and Relaxation Time

The Values of static permittivity (ε_o) and relaxation time (τ) of Ashokarishta – Ethanol system for 11 different concentrations at four temperatures are listed in table 1. The Variation in static permittivity (ε_o) with increasing volume fraction of Ethanol is shown in fig 7. Static permittivity of mixture decreases linearly with increasing volume fraction of Ethanol. Decrease in permittivity with increasing temperature is also observed for all concentrations. This fall in permittivity with increasing amount of solute indicates linear increase in effective dipole moment of the system.

The variation in relaxation time (τ) with increasing volume fraction of Ethanol is shown in fig. 8. Relaxation time increases with increasing volume of Ethanol in solution. Raise in relaxation time (τ) is slow up to 50% Ashokarishta - Ethanol solution. With increase in volume fraction of Ethanol above 50%, relaxation time rises fast. Gradual decrease in relaxation time with increasing temperature is also observed for all concentrations. Fast increase in relaxation time in Ethanol rich region shows that there is significant change in size of molecular structures in this region. The slow increase in relaxation time indicates decrease in density of larger molecular structures in solutions. This increase in relaxation

time indicates increase in amount of hydrogen bonding between solute and solvent molecules, which leads to smaller molecular structures rotating fast.

10.2 Excess permittivity (ε^E) and excess inverse relaxation time $(1/\tau)^E$

The structural changes in binary mixture can explored by determining excess properties. The plot of excess permittivity (ε^E) and excess inverse relaxation time $(1/\tau)^E$ with change in volume. fraction of Ethanol is shown in fig 9 & 10. The values of excess permittivity are positive for all concentrations and at all temperature. The positive values of excess permittivity in mixture indicate increase in effective dipole moments, in proportion to their volume fraction, in pure liquids. This increase in effective dipole moment can be attributed to formation of new smaller structures, may be due to hydrogen bonding, with dipole moment more than addition of dipole moments of constituting molecules. The decrease in relaxation time from value of pure ethanol to 100% Ashokarishta can be explained with breaking of hydrogen bonds in mixture. Further increase in volume fraction of Ashokarishta increases density of comparatively smaller molecules, which leads to decrease in relaxation time. It must be noted that this decrease in relaxation time is rapid which shows formation of smaller structures.

The excess inverse relaxation time $(1/\tau)^E$ reveals speed of rotation of molecular structure. The value of $(1/\tau)^E$ gives us frequency at which dielectric loss is maximum. Positive value of $(1/\tau)^E$ shows increase in frequency at which peak value of dielectric loss occurs.

10.3 Bruggeman Factor

The experimental values together with ideal and theoretical values of Bruggeman of (f_B), plotted against change in volume fraction of Ethanol are shown in fig.11. The values of Bruggeman factor (f_B) of Ashokarishta-Ethanol system for 11 different concentrations at four-temperature are listed in table 2. The Bruggeman mixture formulae state linear relationship between (f_B) and volume fraction of solvent by assuming that there is no interaction between solute and solvent. Modified Bruggeman mixture formula can be used if two components in binary mixture interact. The experimental value of (f_B) for Ashokarishta – Ethanol are fitted to modified Bruggeman mixture formula. When value of numerical fitting parameter "a" is unity, modified Bruggeman mixture formula reduces to original Bruggeman mixture formula. Decrease in value of "a" below unity shows increase in effective volume fraction of solvent in mixture. The small values of "a" indicates significant expansion in effective volume of solvent as well as weak interaction between solute and solvent. Furthermore values of "a" changes remarkably with change in temperature, which shows temperature dependent nature of molecular interactions.

10.4 Thermodynamic Parameters

The variation of conductivity for Ashokarishta with volume fraction of ethanol is shown in Fig. 12. The values of molar enthalpy of activation (ΔH) in KJ/Mole, and Entropy (ΔS) in $J/^0$Kmole, obtained from Eyring's equation are given in table no. 3.

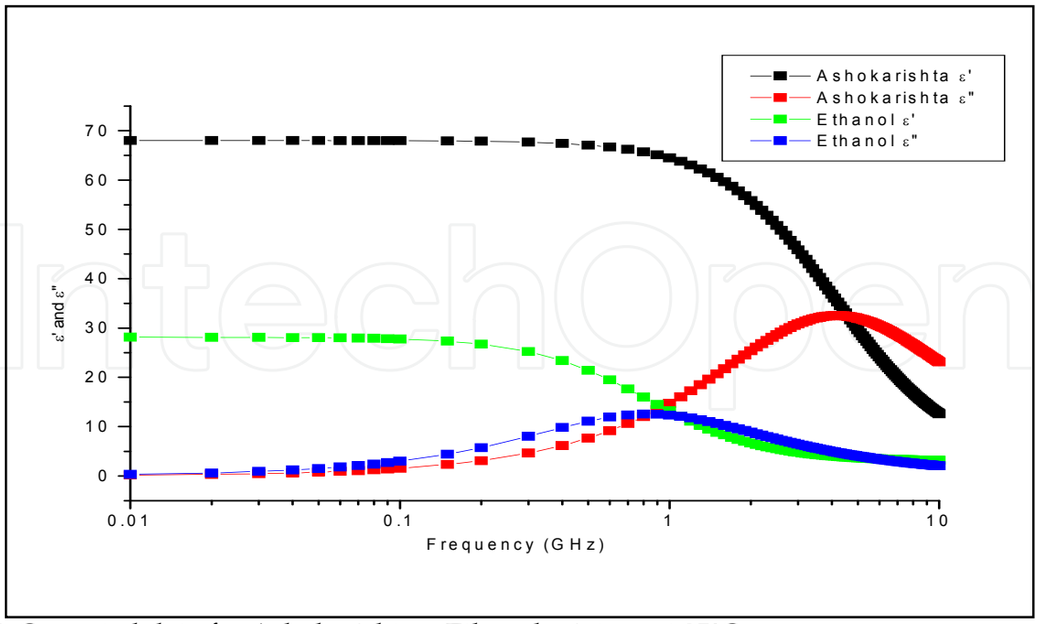

Fig.5. Corrected data for Ashokarishta + Ethanol mixture at 35^0C

The values of (ΔH) and (ΔS) decreases with increases in a volume fraction of Ashokarishta in Ethanol. The value of activation enthalpy gives an idea about nature of compactness in molecules of liquid. The smaller value of (ΔH) shows weaker hydrogen bonding in solute and solvent with decrease in Ashokarishta concentration in mixture. The plot of change in enthalpy and entropy in variation with volume fraction of Ethanol is shown in Fig. 13 and Fig. 14. The variation in free energy of activation with volume fraction of Ethanol in solution is shown in Fig. 15. Arrhenius plot i.e. plot of log (τT) verses 1000/T for Ashokarishta – Ethanol system is shown in the Fig. 16. The nature of plot is almost linear.

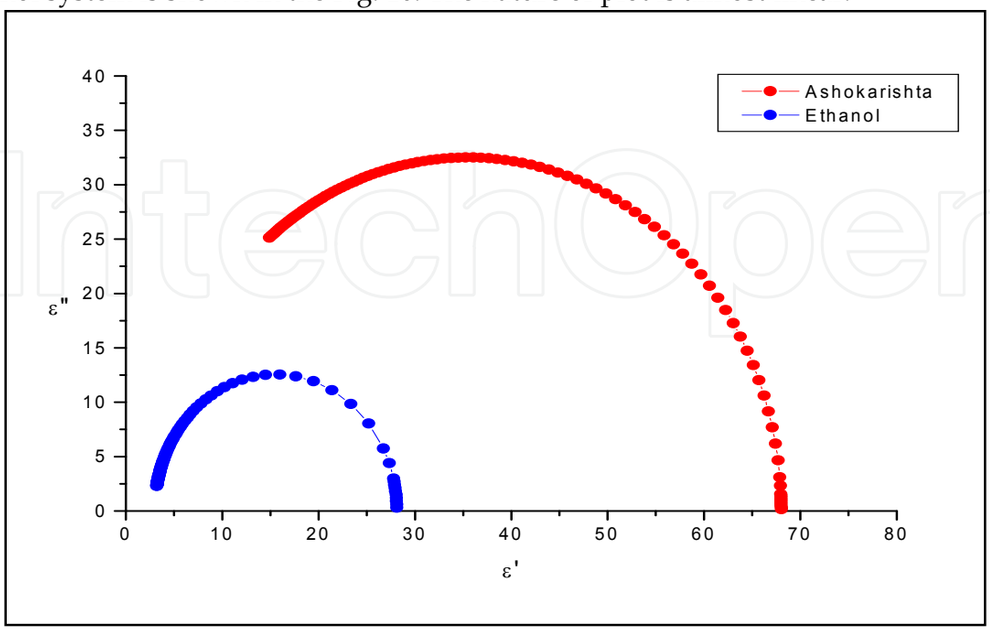

Fig.6. Cole –Cole plot for Ashokarishta + Ethanol

Vol. fraction of Ethanol	Static Dielectric constant (εo)				Relaxation Time (τ)			
	15 ^{0}C	25 ^{0}C	35 ^{0}C	45 ^{0}C	15 ^{0}C	25 ^{0}C	35 ^{0}C	45 ^{0}C
0.0	68.02	65.74	61.18	57.14	38.18	32.18	27.58	25.99
0.1	65.37	62.28	60.82	58.01	43.51	35.62	31.33	26.33
0.2	61.26	58.22	56.74	55.47	49.08	41.66	35.18	31.28
0.3	56.45	54.6	53.2	51.31	53.35	47.54	42.49	31.86
0.4	51.91	50.77	48.95	47.21	58.66	52.98	44.45	36.63
0.5	47.96	46.47	44.54	42.82	64.47	63.67	48.17	37.58
0.6	43.15	41.53	39.29	37.75	73.01	71.45	53.05	46.08
0.7	38.48	39.16	35.66	34.84	83.65	72.55	63.97	57.59
0.8	33.75	32.45	30.98	30.39	98.19	90.30	72.10	66.52
0.9	27.42	27.03	25.61	23.63	149.68	140.89	121.41	90.52
1.0	28.12	26.24	24.94	22.30	194.45	150.75	125.91	108.48

Table 1. Dielectric parameters for Ashokarishta at different temperatures

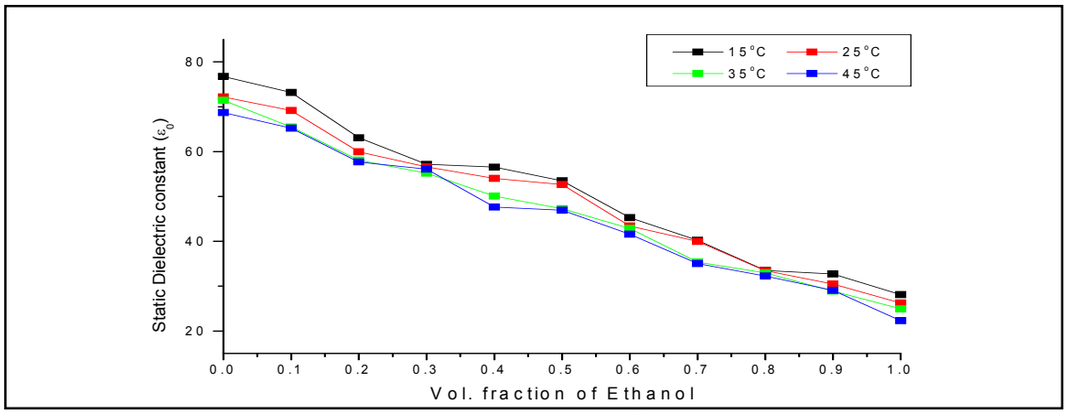

Fig. 7. Variation of permittivity (ε_0) with vol. fraction of Ethanol at various temperatures for Ashokarishta.

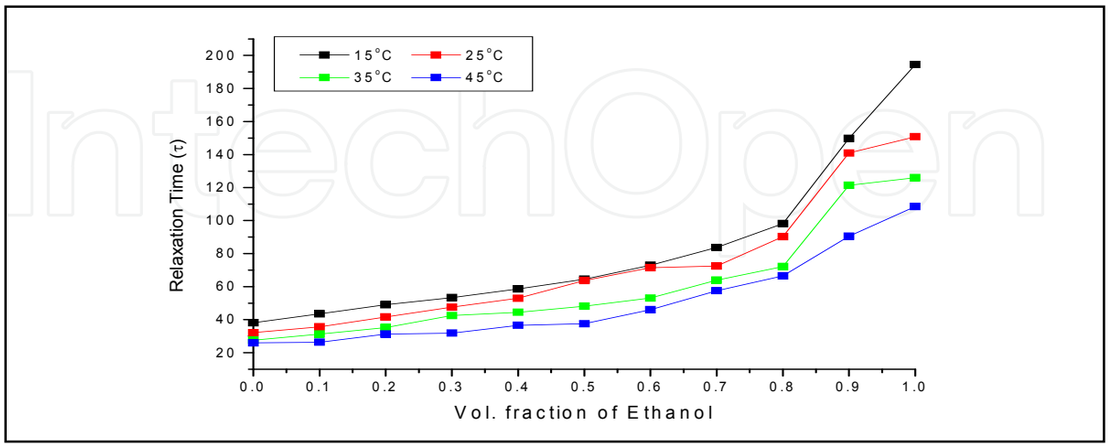

Fig. 8. Variation of relaxation time (τ) with vol. fraction of Ethanol at various temperature for Ashokarishta.

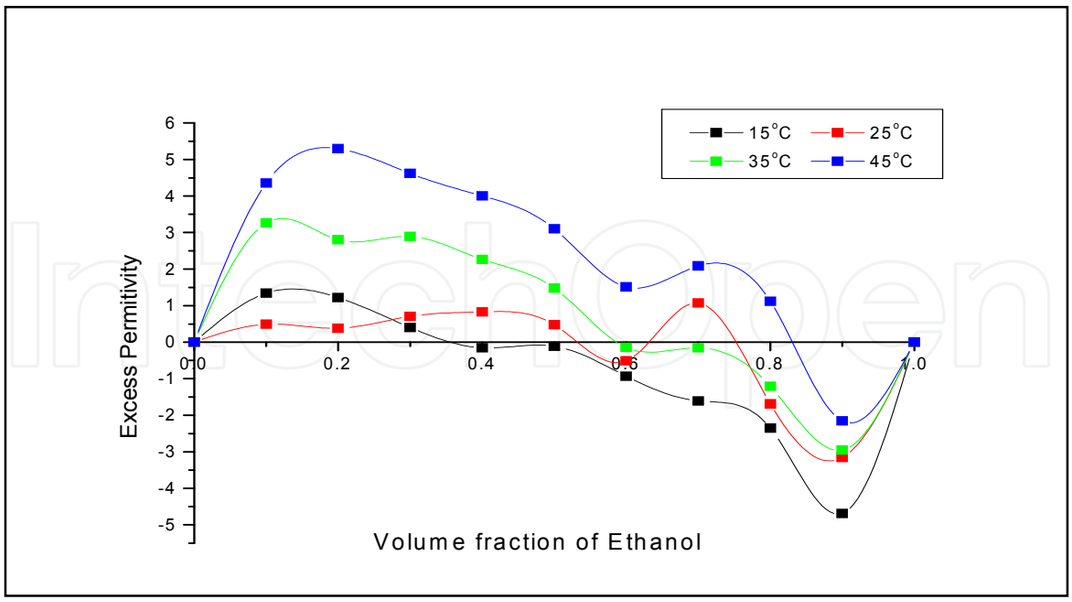

Fig. 9. Variation of excess permittivity (ε^E) with volume fraction of Ethanol in Ashokarishta at various temperatures

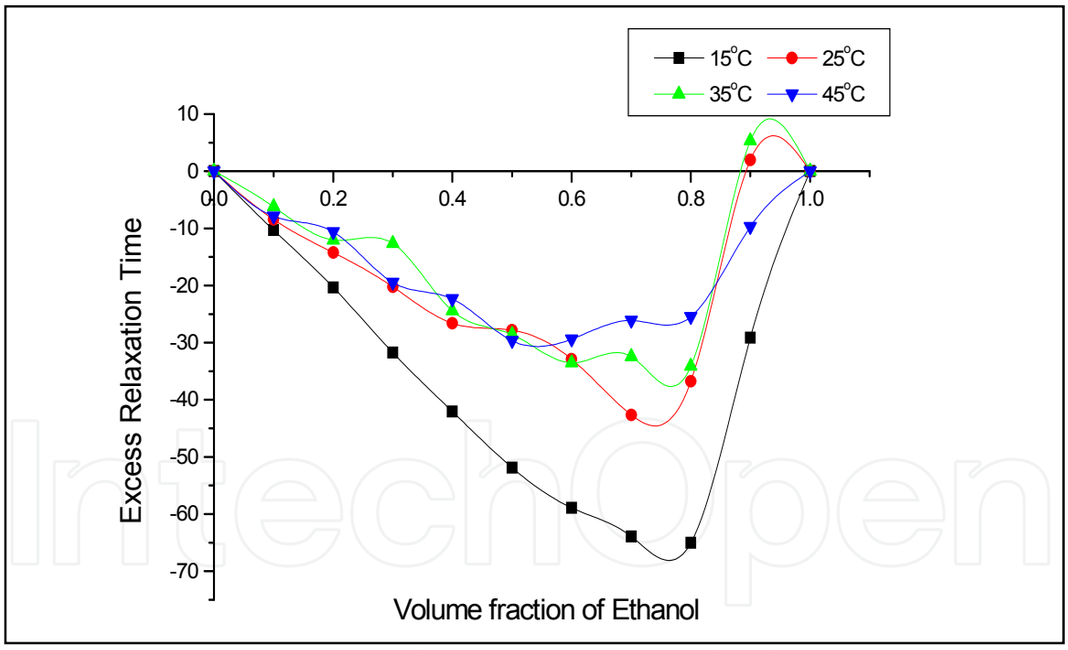

Fig. 10. Variation of excess inverse relaxation time $(1/\tau)^E$ with volume fraction of Ethanol at various temperatures for Ashokarishta.

Vol.fraction of Ethanol	Ideal value for F_b	15 ^{0}C,a=0.933		25 ^{0}C,a=1.0068		35 ^{0}C,a=0.943		45 ^{0}C,a=1.090	
		Expt.	Theor	Expt.	Theor	Expt.	Theor	Expt.	Theor
0.0	1.0	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
0.1	0.9	0.946	0.906	0.929	0.899	0.992	0.905	1.019	0.891
0.2	0.8	0.860	0.810	0.843	0.798	0.899	0.809	0.961	0.785
0.3	0.7	0.755	0.714	0.763	0.698	0.817	0.711	0.863	0.681
0.4	0.6	0.652	0.616	0.676	0.598	0.713	0.613	0.762	0.578
0.5	0.5	0.558	0.516	0.574	0.498	0.601	0.514	0.648	0.477
0.6	0.4	0.438	0.416	0.451	0.398	0.459	0.413	0.509	0.378
0.7	0.3	0.313	0.314	0.388	0.299	0.354	0.311	0.424	0.281
0.8	0.2	0.178	0.210	0.198	0.199	0.209	0.209	0.286	0.185
0.9	0.1	0.023	0.106	0.026	0.100	0.024	0.105	0.051	0.091
1.0	0.0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Table 2. Bruggeman factor for Ashokarishta-Ethanol mixture

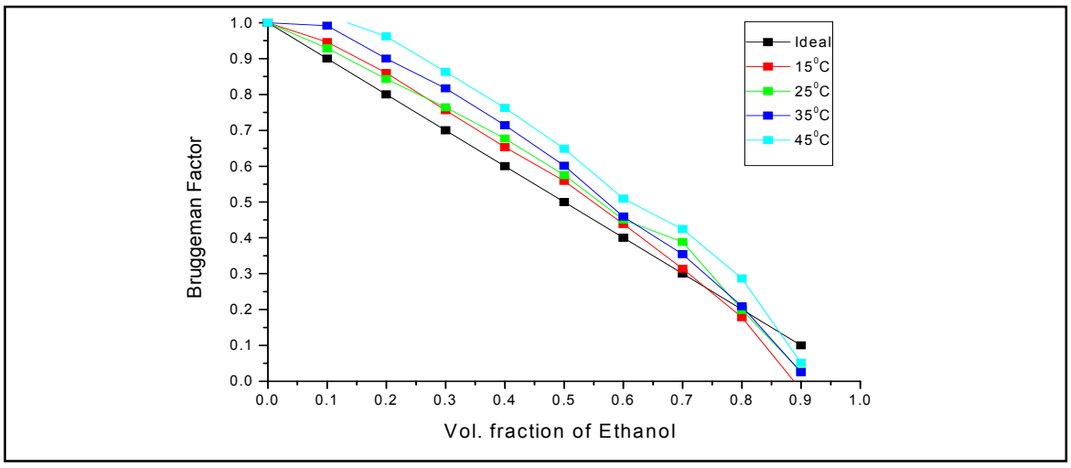

Fig. 11. Variation of Bruggeman factor (F_b) with vol.fraction of Ethanol in Ashokarishta at various temperatures

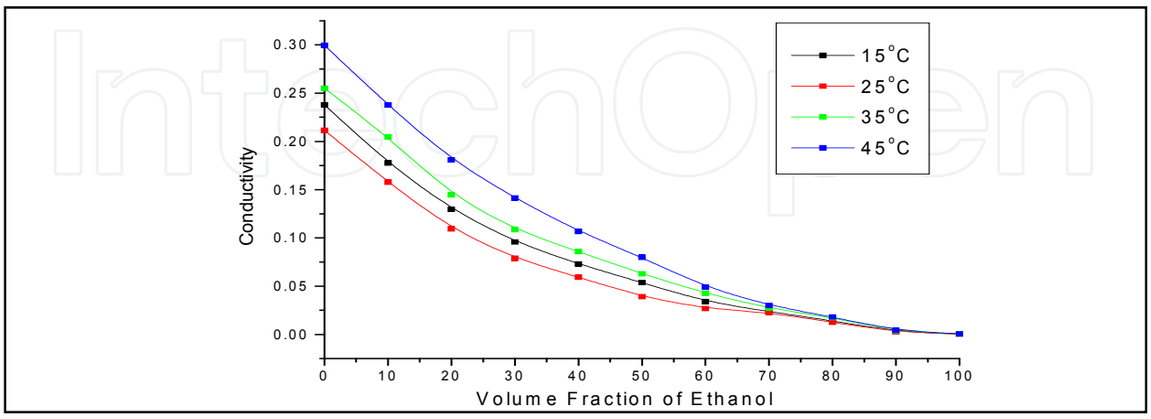

Fig. 12. Variation of conductivity for Ashokarishta with vol.fraction of Ethanol at various temperatures.

Vol. frac. of Ethanol	Entropy (ΔS) J/^{O}Kmole	Enthalpy(ΔH) KJ/Kmole
0.0	-0.019	7.49478
0.1	-0.0116	9.942
0.2	-0.0157	9.07856
0.3	-0.0135	10.0227
0.4	-0.00534	12.64
0.5	-0.00917	11.809
0.6	-0.0156	10.195
0.7	-0.0274	6.9836
0.8	-0.0251	8.08786
0.9	-0.0224	9.9656
1.0	-0.016	12.2479

Table 3. Activation Enthalpy and Entropy of Ashokarishta –Ethanol

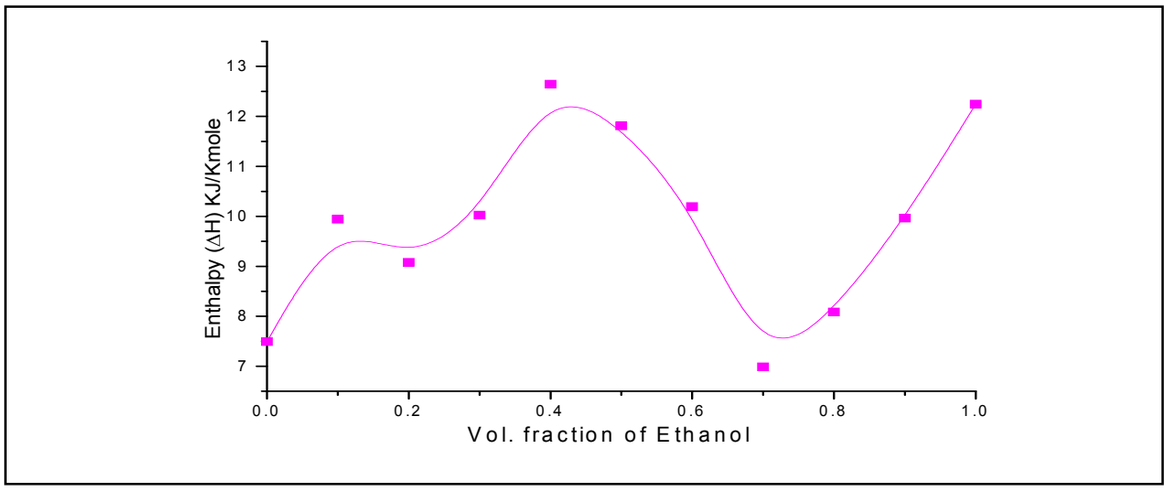

Fig. 13. Variation of Enthalpy for Ashokarishta + Ethanol

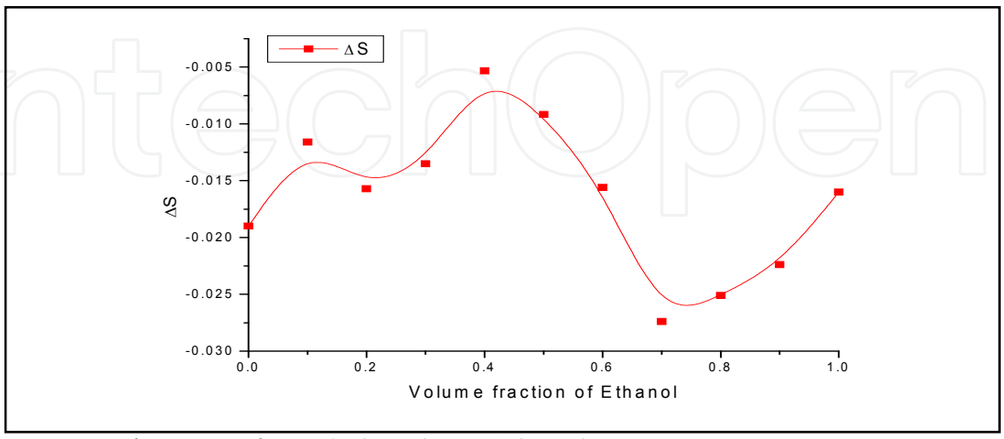

Fig. 14. Variation of Entropy for Ashokarishta + Ethanol

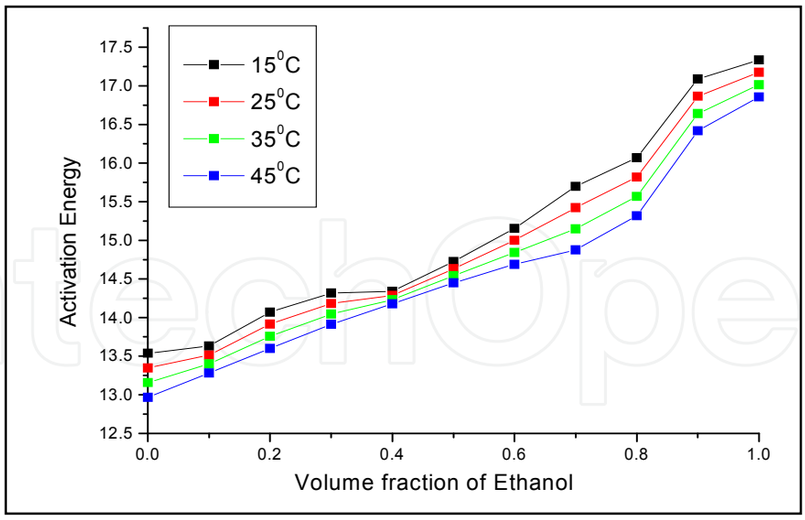

Fig.15. Variation of free energy of activation for Ashokarishta + Ethanol

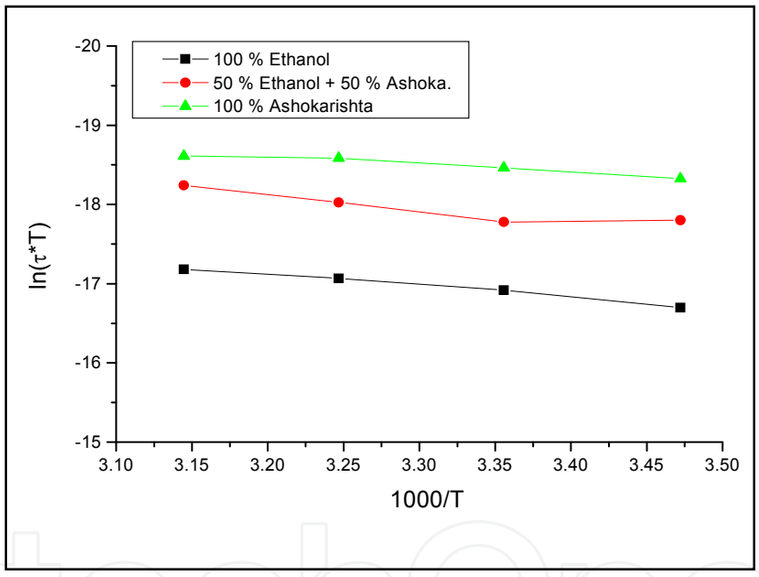

Fig.16. Arrhenius plot of Ashokarishta + Ethanol mixture

11. RESULTS AND DISCUSSION

The temperature dependent dielectric Relaxation as well as frequency dependent dielectric Relaxation has been used to investigate the information of dielectric properties of biological materials. The study of dielectric properties of these materials are of great assistance in exploring the molecular structure and dynamics of condensed matter. We can investigate the information such as molecular flexibility or rigidity, shape and size etc.

When a molecular system is placed in an electric field, there is always the tendency for the electrically charged species to move along the appropriate direction, causing the atom to develop an induced dipole moment. Permittivity of material reflects materials ability to get

polarized with applied electric field. The amount of polarization depends on factors such as size of molecule, effective dipole moment and temperature. In microwave region major contribution to total polarization is orientation polarization. As frequency of applied field increases, it is expected that permittivity should decrease, since molecular orientation cannot cope up with speed with which applied field changes. Hence increase in frequency of applied field decreases alignment of molecular dipoles, which ultimately decrease permittivity. It is very interesting to observe frequency of point from which fall in permittivity begins. This point indicates the beginning of dispersion process. The shift in this point with change in temperature for biological sample gives us an idea about change in induced polarization. Relaxation time of biological material can be related to the size of molecule and mobility of molecules in liquid.

If relaxation time decreases it is correlated that due to decrease in size of molecules as well as to increase in mobility of molecules in liquid. If polar solute molecules are spherical and large by comparison with the solvent molecules then the orientation relaxation of the solute molecules can usefully be described using Debye's model. In this model the dipolar solute molecules are considered as spheres whose rotation is opposed by the viscosity of the surrounding solvent medium.

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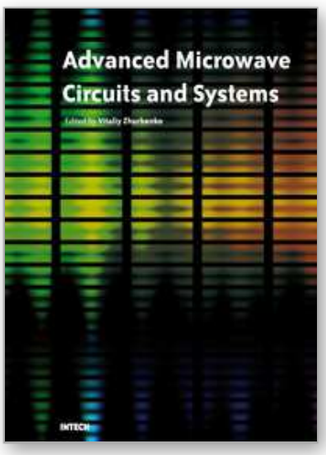

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