Microwave Dielectric characterization of Ayurvedic Medicine using Time Domain Reflectometry Technique

D. P. Chavan¹, C. T. Londhe², A S Disale³ and P B Undre⁴*

¹Department of Botany, Shrikrishna Mahavidyalaya, Gunjoti, Omerga, Maharashtra-413613, India.
²Department of Physics, Mahatma Gandhi Mahavidyalaya, Ahmedpur, Maharashtra-413515, India.
³Vidya Pratishthans Kamalnayan Bajaj Institute of Engineering and Technology, Baramati, District-Pune, India
⁴Microwave Research Laboratory, Department of Physics, Dr. Babasaheb Ambedkar Marathwada University, Aurangabad - 431004, India.

*Corresponding Author Email: prabhakar25u@gmail.com

Abstract. The dielectric dispersion ε’ and dielectric loss ε’’ of ayurvedic medicine such as arjunarishta, khadirarishta and lohasava as were measured by employing the Time Domain Reflectometry technique over a frequency range 10 MHz to 20 GHz at 15, 25, 35 and 45°C. The accuracy in the measurement of the ε’ and ε’’ values obtained from this technique is within ±5%. To evaluate various dielectric parameters, the frequency dependents complex permittivity (ε* (ω) = ε’ - je’’) data viz., dielectric constant, relaxation time and permittivity at high frequency (ε∞) were fitted by the non-linear least-squares fit method to Debye expression. Temperature dependent dielectric constant (ε₀), and relaxation time (τ) have been determined. Further the variation in dielectric parameters and molecular reorientation due to change in the molecular structure has been explored by comparing the values of dielectric parameters of arjunarishta, khadirarishta and lohasava.

1. Introduction
Microwave energy has been directly applied in many fields especially in scientific research such as communication, microwave-assisted chemistry and in food industry [1]. The successful application of microwaves is directly associated with the dielectric properties of the materials. An accurate measurement and working knowledge of these properties are key factors. The knowledge of frequency dependent dielectric properties of solvent systems is important both in fundamental studies of solvent structure and dynamics and in practical application of microwave heating processes [1].

Dielectric relaxation study [2-10] of liquids is one of the most important approaches to understand its molecular behavior. The different types of liquids such as organic liquids [1-10], biological samples [11-12], electrolytes and liquid crystals [13-15] were studied by this method.
To compare the influence of the different polar groups in the relaxation mechanism, we have considered it interesting to perform an analogous treatment such as that carried out for pure and binary mixture of liquids [1-10]. For this reason, dielectric dispersion study in ayurvedic syrup arjunarishta, khadirarishta and lohasava over the frequency range of 10 MHz to 20 GHz have been carried out using pico-second Time Domain Reflectometry. The arjunarishta syrup consists of the natural ingredients such as terminalia arjuna – Stem bark, Mrudvika – Dry grapes, Madhuca Indica – Flower, Woodfodia Fruticosa – Flower, Guda – Jaggery. it is also helpful for febrifuge, cardiac stimulant, cleaning sores and ulcers, keeping digestive system healthy, for the treatment of hemorrhoids, dysentery, diarrhoea, liver diseases, piles, leucorrhoea, menorrhagia, ulcers, wounds, skin diseases, fever, headache, herpes, treatment of several ailments which includes rheumatism, leucorrhoea, menorrhagia, asthma, liver disorder, and inflammatory conditions. Khadirarishta is an ayurvedic blood purifier and an anti-bacterial medicine. It helps in eliminating the toxins and microorganism in the body. It also helps in improving digestion, curing pimples/ acne and other skin related problems such as cysts, gout, herpes, wounds, tumors. Active ingredients present in khadirarishta are Khadir, Deodar, Daruhal di, Trifala, Kankol, Nagkeshar, Jaiphal, Laung, Badi-Elaichi, Dalchini, Pipal, Dhain Ke Phool, Jaggery (Gud), Mishri. Lohasava is an ayurvedic iron tonic as well as anti-obesity medicine. In addition to these benefits, it is also helpful in the management of diseases such as jaundice, hepatitis, spleen enlargement, fatty liver, malabsorption syndrome, cough, asthma etc.

In view of special interest of these ayurvedic syrup, the dielectric relaxation study of arjunarishta, khadirarishta and lohasava at different temperature have been carried out. The aim of this study is to provide the precision dielectric constant and relaxation time values of these ayurvedic syrup and to confirm the nature of H-bonded molecular structures.

2. Experimental Set-Up

2.1. TDR set-up and data acquisition

The Hewlett Packard HP54750A sampling oscilloscope with HP54754A TDR plug in module has been used. After observing TDR response for sample under study, the time window was kept to 5 ns. By observing TDR response for sample under study, the SMA sample cell with 1.35 mm effective pin length has been used. The sample cell holds the liquid under consideration. The physical dimensions of the cell are very important, so one must be careful while designing the sample cell. The impedance of the cell should be matched with coaxial transmission line to which cell is connected. If there is impedance mismatch then unwanted reflections may disturb the wave thereby causing some errors in the measurements. The proper design of cell includes the inner conductor and outer conductor diameters. The length of the inner conductor is called as ‘pin length’ of the cell and is very important factor in analysis. The sample length must be enough to avoid unwanted reflections.

In total reflection method, the sample length must be long enough to produce an adequate difference signal but short enough to keep less complication of resonance effects at frequencies above the range of interest. The characteristics impedance of a coaxial line is given by

\[ Z = \frac{138.2}{\sqrt{\varepsilon}} \log_{10} \frac{b}{a} \]  

(1)

This impedance for our transmission line is frequently 50 Ω. Here ‘a’ is the diameter of inner conductor and ‘b’ is the inner diameter of outer conductor. The \( \varepsilon \) is the relative permittivity of the dielectric between the conductors. Using Teflon and air, and taking appropriate ‘a’, ‘b’ a cell can be designed to have a matching impedance of \( Z = 50 \) Ω (for air \( \varepsilon = 1 \) and for Teflon, \( \varepsilon = 2.2 \)). The sample cell consists of standard military applications (SMA) coaxial connector with matched impedance with 3.5 mm inner diameter of outer conductor. The inner conductor of SMA connector itself is considered as ‘inner conductor’ and hex-nut is treated as an outer conductor. Since these SMA connectors have already been designed for precise 50 Ω impedance, a special design, when used with high frequency,
is not required. The physical length of inner conductor can be changed. When cell is filled with sample above the physical length of inner conductor the fringing effect takes place [16,17]. Due to the fringing field the effective pin length [18] will not be equal to physical pin length. The effective electrical pin length will be more than the physical pin length. The accurate determination of effective pin length ‘d’ is very important for the accurate evaluation of dielectric parameters. It is found that for SMA type cell, effective pin length [18] is greater than actual physical length by 0.1 - 0.2 mm.

To reduce noise, time dependent response curve was averaged for 64 times and then stored in TDR oscilloscope memory with 1024 points per waveform. First, the response waveform for empty cell is acquired and stored in memory and then secondly, the response waveform for sample is acquired and stored in other memory. The empty cell waveform is used as reference waveform. Both the response waveforms are the reflected waveforms from the sample cell with open termination transmission line.

The data acquisition is carried out for 11 concentrations at 15, 25, 35 and 45°C. The temperature of sample was maintained at desired value, within accuracy limit of ± 1°C, by circulating constant temperature water through heat insulating jacket surrounding sample cell.

At each time the response waveforms without sample and with sample were recorded. The time dependent response waveform without sample is referred as $R_1(t)$ and with sample referred as $R_x(t)$.

2.2. Data analysis
The time dependent data were processed to obtain complex reflection coefficient spectra $\rho^*(\omega)$ over the frequency range from 10 MHz to 10 GHz using Fourier transformation as [19,20]

$$\rho^*(\omega) = \left( \frac{c}{j\omega d} \right) \left[ \frac{p(\omega)}{q(\omega)} \right]$$

(2)

Where $p(\omega)$ and $q(\omega)$ are Fourier transformations of $(R_1(t) - R_x(t))$ and $(R_1(t) + R_x(t))$, respectively, $c$ is the velocity of light, $\omega$ is angular frequency, $d$ is effective pin length and $j = \sqrt{-1}$. The complex permittivity spectra $\varepsilon^*(\omega)$ [21] were obtained from reflection coefficient spectra $\rho^*(\omega)$ by applying bilinear calibration method [22]. The complex permittivity spectra measured using TDR are fitted by the non-linear least square fit method to the Havriliak Negami expression [23] to obtain various dielectric parameters. The accuracy in the measurement of dielectric dispersion $\varepsilon'$ and dielectric loss $\varepsilon''$ values obtained from this technique is within ±5%.

$$\varepsilon'(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)}{[1+(\omega\tau)^{1-\alpha\beta}]}$$

(3)

where $\varepsilon^*(\omega)$ is the complex permittivity at an angular frequency $\omega$, $\varepsilon_\infty$ is the permittivity at high frequency, $\varepsilon_0$ is the static permittivity, $\tau$ is the relaxation time of the system, $\alpha$ is the shape parameter representing symmetrical distribution of relaxation time and $\beta$ is the shape parameter of an asymmetric relaxation curve. Equation (3) includes Cole-Cole ($\beta = 1$) [24] Davidson-Cole ($\alpha = 0$) [25] and Debye ($\alpha = 0, \beta = 1$) [26] relaxation models. The dielectric model for fitting dielectric parameters suitable for present system is Debye dispersion model.

3. Results and Discussions:
Frequency dependent permittivity of materials reflects materials ability to get polarized with applied electric field. In microwave major contribution to total polarization is orientation polarization. The amount of polarization depends on factors such as size of molecules, effective dipole moment and temperature. In general fall in permittivity with increase in frequency of applied field is expected since molecular orientation cannot cop-up with the speed with which applied field changes. Thus, increase in frequency of applied field decreases alignment of molecular dipoles, which ultimately decreases permittivity. It is very interesting to observe frequency at a 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 pure liquid gives us an idea about change in induced polarization. Relaxation time of material can be related to the size of molecule and to the mobility of molecules in liquid. Decrease in relaxation time can be correlated to decrease in size of molecules as well as increase in mobility of molecules in liquid.

![Graph showing dielectric permittivity and loss for different temperatures](image1)

**Figure 1.** Frequency dependent dielectric permittivity/dispersion ($\varepsilon'$) and dielectric loss ($\varepsilon''$) curves for arjunarishta syrup at 15, 25, 35 and 45°C.

![Graph showing dielectric permittivity and loss for different temperatures](image2)

**Figure 2.** Frequency dependent dielectric permittivity/dispersion ($\varepsilon'$) and dielectric loss ($\varepsilon''$) curves for khadirarishta syrup at 15, 25, 35 and 45°C.
Figure 3. Frequency dependent dielectric permittivity/dispersion ($\varepsilon'$) and dielectric loss ($\varepsilon''$) curves for lohasava syrup at 15, 25, 35 and 45°C.

Frequency dependent curves for the dielectric permittivity / dispersion ($\varepsilon'$) and absorption ($\varepsilon''$) for arjunarishta, khadirarishta and lohasava syrup are depicted in figure 1, 2 and 3, respectively. Measurements of the complex permittivity i.e. dielectric dispersion ($\varepsilon'$) and absorption ($\varepsilon''$) were carried out over a frequency range 10 MHz-10 GHz by employing the TDR at different four temperatures. The maximum errors in the evaluated values of $\varepsilon'$ and $\varepsilon''$ are ±2 and ±3%, respectively. It has been found that the complex dielectric data of arjunarishta, khadirarishta and lohasava systems obey the Debye dispersion model faithfully.

It is observed from the permittivity curves that fall in permittivity starts at higher frequency with increasing temperature. This indicates better alignment of molecular dipoles, which needs higher frequency of applied field to get disturbed. Dielectric loss is the amount of incident energy dissipated in form of heat. Increase in temperature of liquid decreases this loss. This can be observed from decrease in peak value of dielectric loss with increase in temperature. The broadening of dispersion curves with increase in temperature indicated better alignment of dipoles with field. The decrease in dielectric loss with increase in temperature indicates more ordered arrangements of dipoles. Temperature dependent dielectric loss peaks for arjunarishta, khadirarishta and lohasava molecules are found in the frequency range from approximately 4.8 to 7.6 GHz, 5.6 to 8.3 GHz, 5.5 to 9.5 GHz, respectively.

From Figure 1, 2 and 3, it is observed that the relaxation time is shorter for lohasava compared with arjunarishta and khadirarishta. On the other hand, the temperature dependence of the dielectric constant (Figure 4) and of the relaxation time (Figure 5) is stronger in arjunarishta compared with khadirarishta and lohasava, as a consequence of the reported differences between these liquids. Moreover, the dielectric strength of the relaxation is in the order arjunarishta $>$ khadirarishta $>$ lohasava, which indicates that the relaxing dipole moment is higher in arjunarishta.
Figure 4. Variation in dielectric constant ($\varepsilon_0$) with temperature for arjunarishta, khadirarishta and lohasava.

From figure 4, it is observed that dielectric constant values decrease with increase in temperature for all ayurvedic syrup. This loosely bound but with highly correlated system of dipoles arranges itself more regularly which needs higher frequency of applied field to get disturbed. From Figure 5, it is found that the $\tau$ values also decrease with increase in temperature for all ayurvedic syrup. This decrease in relaxation time reveals fast rotation of molecules, which may be due to increase in mobility of molecules with increase in temperature. The temperature dependent $\tau$ values of arjunarishta, khadirarishta and lohasava shows that the decrease in $\tau$ of lohasava is much higher in comparison to the $\tau$ values of arjunarishta and khadirarishta with rise in temperature. This suggests
that there is a large decrease in the hindrance to the molecular reorientation for lohasava molecules with increase in temperature.

4. Conclusions
The dielectric dispersion $\varepsilon'$ and dielectric loss $\varepsilon''$ of ayurvedic syrup such as arjunarishta, khadirarishta and lohasava have been studied using TDR technique in the frequency range 10 MHz to 20 GHz over entire concentration at 15, 25, 35 and 45°C. The dielectric dispersion behavior of all three ayurvedic syrup under study is found to be Debye type. It is observed from the permittivity curves that fall in permittivity starts at higher frequency with increasing temperature. The dielectric constant and relaxation time values are found in the order arjunarishta > khadirarishta > lohasava at all four temperature.

References

[1] Undre P B, Khirade P W and Mehrotra S C, 2012 Microwave dielectric characterization of 1, 2-diaminopropane, 1, 3-diaminopropane and 3, 3'-diaminodipropylamine Asian Journal of Spectroscopy pp 199-209

[2] Undre P B, Khirade P W, Jagdale S B, Helambe S N and Mehrotra S C, 2011 Dielectric studies on binary mixtures of formamide with ethanolamine using the time domain technique Lithuanian Journal of Physics 51(2) pp 147-154

[3] Undre P B, Patil S S and Khirade P W, 2013 Dielectric characterization and molecular interaction behaviour in binary mixture of 1, 2-diaminopropane with 2-aminoethanol Main Group Chemistry 12(4) pp 361-373

[4] Undre P, Helambe S N, Jagdale S B, Khirade P W and Mehrotra S C, 2007 Microwave dielectric characterization of binary mixture of formamide with N, N-dimethylaminoethanol Pramana 68(5) pp 851-861

[5] Undre P, Helambe S N, Jagdale S B, Khirade P W and Mehrotra S C, 2008 Study of solute–solvent interaction through dielectrics properties of N, N-dimethylacetamide in ethanolamine Journal of Molecular Liquids, 137(1-3) pp 147-151

[6] Undre P B, Deshmukh M L, Londhe C T and Khirade P W, 2020 Microwave Dielectric Relaxation in Binary Mixtures of 1, 3-Diaminopropane in Dimethylaminoethanol Integrated Ferroelectrics 205(1) pp 164-176

[7] Maheshmakar P R, Sayyad S B, Shivalkar K N, Kamble S P, Undre P B and Khirade P W, 2012 Dielectric Relaxation Study of Diethylene Glycol Monomethyl Ether with Aromatic Compounds Using Time Domain Reflectometry Technique Asian Journal of Chemistry 24(12) pp 5711-5714

[8] Jadhavpatil V, Undre P and Helambe S, 2019 Dielectric Relaxation in Water-Ethanolamine Mixtures as a Function of Composition and Temperature Integrated Ferroelectrics 202(1) pp 112-121

[9] Kolhe S B, Undre P B, Deshpande V P and Khirade P W, 2019 Dielectric characterization and molecular interaction behaviour in binary mixtures of methyl acetate with 1-butanol and 1-pentanol Indian Journal of Pure and Applied Physics 57 pp 900-910

[10] Disale A S, Undre P B, Yaseen S A, Saif F A, Alameen A S, Patil S S and Khirade P W, 2019 Dielectric Relaxation and FTIR Studies on Molecular Interaction between Ethylene Glycol Monobutyl Ether with Bromobenzene and Chlorobenzene Integrated Ferroelectrics 202(1) pp 67-78

[11] Ranade A A, Undre P B, Barpande S R, Tukpvari J V and Mehrotra S C, 2016 Salivary dielectric properties in oral cancer (OSCC) through time domain reflectometry at microwave region: the future alternative for diagnosis and treatment Global Journal of Medical Research: F Diseases 16(1) p 13

[12] Ranade A A, Undre P B, Barpande S R, Tukpvari J V, Mehrotra S C, 2015 Microwaves and mobile waves: Future of oral cancer diagnosis and treatment International Journal of
Preventive & Clinical Dental Research 2(4) pp 26-30
[13] Kalaivani T, Undre P, Sabesan R and Krishnan S, 2012 Dielectric relaxation studies of nitriles solubilized by Sodium dodecyl sulphate in aqueous solutions Journal of Molecular Liquids 172 pp 76-80
[14] Kalaivani T, Undre P, Sabesan R and Krishnan S, 2009 Dielectric relaxation studies of aqueous sodium dodecyl sulfate with some amines as co-solvents by time domain reflectometry technique Main Group Chemistry 8(2) pp 125-141
[15] Kalaivani T, Undre P, Sabesan R and Krishnan S, 2009 Dielectric relaxation studies of aqueous sodium dodecyl sulfate with some amines as co-solvents by time domain reflectometry technique Main Group Chemistry 8(2) pp 125-141
[16] van Gemert M C, 1974 Theoretical analysis of the lumped capacitance method in dielectric time domain spectroscopy Advances in Molecular Relaxation Processes 6(2) pp 123-137
[17] d’Anna G and Benoit W, 1990 Apparatus for dynamic and static measurements of mechanical properties of solids and of flux-lattice in type-II superconductors at low frequency (10−5–10 Hz) and temperature (4.7–500 K) Review of scientific instruments 61(12) pp 3821-3826.
[18] Berberian J G and King E, 2002 An overview of time domain spectroscopy Journal of non-crystalline solids 305(1-3) pp 10-18.
[19] Shannon C E, 1949 Communication in the presence of noise Proceedings of the IRE 37(1) pp 10-21.
[20] Samulon H A, 1951 Spectrum analysis of transient response curves Proceedings of the IRE 39(2) pp 175-186.
[21] Mashimo S, Kuwabara S, Yagihara S and Higasi K, 1989 The dielectric relaxation of mixtures of water and primary alcohol The Journal of chemical physics 90(6) pp 3292-3294.
[22] Cole R H, Berberian J G, Mashimo S, Chryssikos G, Burns A and Tombari E, 1989 Time domain reflection methods for dielectric measurements to 10 GHz Journal of applied physics 66(2) pp 793-802.
[23] Havriliiak S and Negami S, 1966 A complex plane analysis of α-dispersions in some polymer systems Journal of Polymer Science Part C: Polymer Symposia 14(1) pp 99-117.
[24] Cole K S and Cole R H, 1941 Dispersion and absorption in dielectrics I. Alternating current characteristics Journal of chemical physics 9(4) pp 341-351.
[25] Davidson D W and Cole R H, 1950 Dielectric relaxation in glycerine Journal of Chemical Physics 18(10) p 1417
[26] Debye P, 1929 Polar Molecules (Chemical Catalog Company Inc, New York).