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Frequency and Temperature Dependent Dielectric Relaxation Studies of Paracetamol in the Dilute Solution of Carbon Tetrachloride

Chitra Manro[1], Ritu Jain[2]

[1]Department of Physics, Swami Keshvanand Institute of Technology Management & Gramothan, Ramnagaria, Jaipur, Rajasthan, India

[2]Department of Physics, The IIS University, Gurukul Marg, SFS, Mansarovar, Jaipur, Rajasthan, India

***Abstract*— The dielectric relaxation time (τ) and the dipole moment (μ) of Paracetamol are measured in the dilute solution of carbon tetrachloride for different concentrations at three different frequencies 9.265 GHz, 7.275 GHz and 4.64 GHz at four different temperatures (303K, 313K, 323K and 333K). Different dielectric parameters like dielectric constant (ε'), dielectric loss factor (ε''), static permittivity (ε_0) and dielectric constant at optical frequencies (ε_∞) have been calculated by using standard methods. Dielectric constant (ε') and dielectric loss factor (ε'') are observed to decrease with temperature and increase with frequency, whereas the values of dielectric relaxation time are found to decrease with temperature and frequency. Dipole moment values are decreasing with temperature but they are independent of frequency.**

***Keywords*— Dielectric Parameters, Dielectric Relaxation Time, Dipole Moment**.

I. INTRODUCTION

Paracetamol is a popular and common drug[1]around the world. It is anenalgesic and antipyretic drug[2] whose production is very large. Its chemical formula is $CH_3CONHC_6H_4OH$. It is available in the market with or without prescription. In the present paper we are studying the dielectric properties of paracetamol which is dissolved in the dilute solution of carbon tetrachloride. These properties are useful to provide important information about the complex formation in solution[3] and also about the strength and nature of intermolecular interaction.[4] These studies also help to understand

Kumar et al.[5] calculated the dielectric relaxation and dipole moment of binary mixtures of ethylene glycol, propylene glycol and butylene glycol with dilute solutions of 1, 4-Dioxane for different concentrations at 306K. The static dielectric constants (ε_0) of glycols, like ethylene glycol, propylene glycol and butylene glycol in dilute solutions of 1,4-Dioxane were also determined at 303K.

The measuring frequency of the dipole meter used by them was 2MHz. X-band and J-band microwave benches operating at 9.52GHz and 7.72GHz were used for the determination of dielectric permittivity (ε') and dielectric loss factor (ε''). The values of molecular relaxation time (τ_0) and dipole moment (μ) for different compositions of binary mixtures were determined using Higasi's method. They concluded that the relaxation time values increase with increasing viscosity of the medium.

The studies of dielectric properties of polar liquids, especially in dilute solutions with non-polar medium have important role in liquid state. Gedam et al.[6] concluded that the dielectric investigations mainly probe weak forces between the molecules and help to understand intermolecular reorientation dynamics of the solute. The values of dielectric constant (ε') and dielectric loss (ε'') of polar liquids in dilute solution of benzene increase as a function of concentration of polar substance.Sahoo et al.[7] observed that the simple Debye model of polar and non polar liquids can satisfactorily explain the dielectric behavior of amides and acetone under static and high frequency electric fields. By using the values of ε_0 and ε_∞ we can further calculate different relaxation times. Thus the relaxation studies can provide useful information about the nature of molecular orientation process.[8]

II. EXPERIMENTAL DETAILS

In the present investigation, different mole fractions of Paracetamol (0.01, 0.015, 0.02, 0.025, 0.03) are dissolved in 1 mole of carbon tetrachloride to make dilute solution samples. Here CCl_4 used as a solvent is procured from Merck and AR grade Paracetamol was purchased from the Central Drug House, Delhi, India. The values of dielectric constant and dielectric loss for the samples are determined at three different microwave frequencies viz 9.265 GHz, 7.275 GHz and 4.64GHz by using X-band, J-band and C-band microwave benches respectively at four different temperatures (303K, 313K, 323K and 333K).

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The values of dielectric permittivity at low frequencies (ε_0) and at optical frequencies (ε_∞) are determined by using a dipole meter[9] and Abbe's refractometer[10] respectively. All the measurements are performed at different temperatures using a constant temperature circulating water bath fitted with a thermostat having temperature stability of the order of ± 0.1°C.

For dilute solutions in non-polar solvents ε', ε'', ε_0 and ε_∞ can be expressed as linear functions of concentrations[11-12] in the following manner:

$$\varepsilon' = \varepsilon'_1 + a'W_2 \quad (1)$$

$$\varepsilon'' = a''W_2 \quad (2)$$

$$\varepsilon_o = \varepsilon_{1o} + a_o W_2 \quad (3)$$

$$\varepsilon_\infty = \varepsilon_{1\infty} + a_\infty W_2 \quad (4)$$

Here subscript 1 refers to the pure solvent, 2 to the solute, while 0 refers to the static or low frequency case and ∞ refers to the infinite or optical frequency case, W_2 is taken as the weight fraction of the solute. a', a'', a_0 and a_∞ are the slopes of above mentioned linear equations. The values of the relaxation times τ_0, τ_1, and τ_2 are calculated by using the method given by Higasi et al[13]. The relaxation time for overall molecular rotation (τ_1) is defined by

$$\tau_1 = \frac{1}{\omega}\left(\frac{a''}{a' - a_\infty}\right) \quad (5)$$

Whereas the relaxation time for intermolecular rotations (τ_2) is given by

$$\tau_2 = \frac{1}{\omega}\left(\frac{a_o - a'}{a''}\right) \quad (6)$$

Here ω is the angular frequency corresponding to the microwave frequency at which the experiment is performed. The most probable relaxation time (τ_0) is then obtained by employing the following relation:

$$\tau_0 = \frac{1}{\omega}\left(\frac{A^2 + B^2}{C^2}\right)^{\frac{1}{2(1-\alpha)}} \quad (7)$$

where

$$1 - \alpha = \frac{2}{\pi}\tan^{-1}\frac{A}{B} \quad (8)$$

and

$$A = a''(a_o - a_\infty)$$

$$B = (a_o - a')(a' - a_\infty) - (a'')^2$$

$$C = (a' - a_\infty)^2 (a'')^2$$

The value of the dipole moment (μ) of the solute molecules is calculated by using Higasi's method. According to this method the value of dipole moment is given by

$$\mu = \left(\frac{27KTM_2}{4\pi N d_1 (\varepsilon_{01} + 2)^2}\right)^{\frac{1}{2}} (a_o - a_\infty)^{\frac{1}{2}} \quad (9)$$

Where M_2 is the molecular weight of solute, d_1 is the density of solvent, k is the Boltzmann constant, N is the Avogadro's number and T is the temperature in Kelvin at which the experiment is performed. Thus knowing the value of a_0 and a_∞, μ can be obtained from eq. (9) at different temperatures.

III. Result And Discussion

In the present paper, we have found the dielectric parameters (ε', ε'', ε_0 and ε_∞) of paracetamol for different mole fractions (0.01, 0.015, 0.02, 0.025 and 0.03) at four different temperatures when it is dissolved in 1 mole of CCl_4. The dielectric constant and dielectric loss are calculated at different frequencies by using X-band, J-band and C-band microwave benches. By using Abbe's refractometer and Dipole Meter the values of optical and static permittivity are calculated which are mentioned in Tables I, II and III for all the three benches.

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Table I
The Values Of Different Dielectric Parameters Like Dielectric Constant (ε'), Dielectric Loss Factor (ε''), Static Permittivity (ε_0) and Dielectric Constant at Optical Frequencies (ε_∞), Relaxation Times and Dipole Moments at Four Different Temperature Using C-Band Microwave Bench. The Values of Relaxation Times in Pico-Seconds and Dipole Moment is in Debey.

Temp. 303 K.								
W.Fr.	**ϵ'**	**ϵ''**	**ϵ_0**	**ϵ_∞**	**τ_0**	**τ_1**	**τ_2**	**μ**
0.00919	2.100	.0240	2.292	2.096	5.2	4.9	5.4	10.03
0.01373	2.101	.0241	2.313	2.098				
0.01823	2.102	.0275	2.345	2.099				
0.02268	2.103	.0276	2.384	2.101				
0.02710	2.104	.0277	2.405	2.102				
Temp. 313 K.								
W.Fr.	**ϵ'**	**ϵ''**	**ϵ_0**	**ϵ_∞**	**τ_0**	**τ_1**	**τ_2**	**μ**
0.00919	2.099	.0206	2.257	2.088	4.7	4.3	5.2	9.85
0.01373	2.100	.0207	2.304	2.089				
0.01823	2.101	.0240	2.318	2.090				
0.02268	2.102	.0241	2.323	2.092				
0.02710	2.103	.0275	2.340	2.093				
Temp. 323 K.								
W.Fr.	**ϵ'**	**ϵ''**	**ϵ_0**	**ϵ_∞**	**τ_0**	**τ_1**	**τ_2**	**μ**
0.00919	2.098	.0171	2.254	2.080	3.8	3.3	4.4	9.48
0.01373	2.099	.0206	2.301	2.082				
0.01823	2.100	.0239	2.306	2.083				
0.02268	2.101	.0240	2.311	2.085				
0.02710	2.102	.0274	2.336	2.087				
Temp. 333 K.								
W.Fr.	**ϵ'**	**ϵ''**	**ϵ_0**	**ϵ_∞**	**τ_0**	**τ_1**	**τ_2**	**μ**
0.00919	2.097	.0137	2.250	2.063	2.6	1.8	3.8	9.42
0.01373	2.098	.0172	2.293	2.064				
0.01823	2.099	.0206	2.298	2.066				
0.02268	2.100	.0238	2.302	2.066				
0.02710	2.101	.0241	2.318	2.069				

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Table II
The Values Of Different Dielectric Parameters Like Dielectric Constant (ε'), Dielectric Loss Factor (ε''), Static Permittivity (ε_0) and Dielectric Constant at Optical Frequencies (ε_∞), Relaxation Times and Dipole Moments at Four Different Temperature Using J-Band Microwave Bench. The Values of Relaxation Times in Pico-Seconds and Dipole Moment is in Debey.

Temp. 303 K.								
W. Fr.	ϵ'	ϵ''	ϵ_0	ϵ_∞	τ_0	τ_1	τ_2	**μ**
0.00919	2.215	0.052	2.292	2.096	3.5	3.2	3.9	10.03
0.01373	2.224	0.053	2.313	2.098				
0.01823	2.240	0.054	2.345	2.099				
0.02268	2.255	0.055	2.384	2.101				
0.02710	2.270	0.057	2.405	2.102				
Temp. 313 K.								
W. Fr.	ϵ'	ϵ''	ϵ_0	ϵ_∞	τ_0	τ_1	τ_2	**μ**
0.00919	2.199	0.045	2.257	2.088	2.9	2.3	3.7	9.85
0.01373	2.210	0.049	2.304	2.089				
0.01823	2.226	0.052	2.318	2.090				
0.02268	2.239	0.053	2.323	2.092				
0.02710	2.256	0.055	2.340	2.093				
Temp. 323 K.								
W. Fr.	ϵ'	ϵ''	ϵ_0	ϵ_∞	τ_0	τ_1	τ_2	**μ**
0.00919	2.196	0.042	2.254	2.080	2.1	1.3	3.3	9.48
0.01373	2.203	0.045	2.301	2.082				
0.01823	2.215	0.048	2.306	2.083				
0.02268	2.230	0.052	2.311	2.085				
0.02710	2.241	0.053	2.336	2.087				
Temp. 333 K.								
W. Fr.	ϵ'	ϵ''	ϵ_0	ϵ_∞	τ_0	τ_1	τ_2	**μ**
0.00919	2.189	0.039	2.250	2.063	1.5	1.1	2.2	9.42
0.01373	2.199	0.041	2.293	2.064				
0.01823	2.212	0.045	2.298	2.066				
0.02268	2.225	0.051	2.302	2.066				
0.02710	2.361	0.052	2.318	2.069				

Table III
The Values Of Different Dielectric Parameters Like Dielectric Constant (ε'), Dielectric Loss Factor (ε''), Static Permittivity (ε_0) and Dielectric Constant at Optical Frequencies (ε_∞), Relaxation Times and Dipole Moments at Four Different Temperature Using X-Band Microwave Bench. The Values of Relaxation Times in Pico-Seconds and Dipole Moment is in Debey.

W. Fr.	€'	€''	$€_0$	$€_\infty$	τ_0	τ_1	τ_2	μ
Temp. 303 K.								
0.00919	2.282	0.197	2.292	2.096	1.5	1.3	1.7	10.03
0.01373	2.306	0.201	2.313	2.098				
0.01823	2.331	0.211	2.345	2.099				
0.02268	2.346	0.224	2.384	2.101				
0.02710	2.368	0.228	2.405	2.102				
Temp. 313 K.								
0.00919	2.188	0.181	2.257	2.088	1.2	.97	1.4	9.85
0.01373	2.199	0.183	2.304	2.089				
0.01823	2.229	0.191	2.318	2.090				
0.02268	2.269	0.194	2.323	2.092				
0.02710	2.313	0.201	2.340	2.093				
Temp. 323 K.								
0.00919	2.181	0.164	2.254	2.080	.98	.92	1.1	9.48
0.01373	2.194	0.180	2.301	2.082				
0.01823	2.205	0.181	2.306	2.083				
0.02268	2.248	0.183	2.311	2.085				
0.02710	2.295	0.191	2.336	2.087				
Temp. 333 K.								
0.00919	2.133	0.156	2.250	2.063	.88	.77	.96	9.42
0.01373	2.134	0.167	2.293	2.064				
0.01823	2.143	0.168	2.298	2.066				
0.02268	2.172	0.179	2.302	2.066				
0.02710	2.209	0.189	2.318	2.069				

From the tables it is clear that the values of dielectric parameters increase linearly with the weight fraction of the solute. This behavior suggests that there is no change in the nature[10] of the rotating molecular entities in the CCl_4 solutions. This may occur due to the dipole-dipole[14] interaction of pure liquid. This may also be due to the increase[15] in the molar volume of molecules.

Table I, II and III also show that the values of all the dielectric parameters decrease as the temperature increases for a particular weight fraction. It indicates the existence of inter molecular and intra molecular association in the solution. To minimize the solute-solute interaction the concentration of the solutions were made sufficiently dilute[6].

The measured values of different dielectric relaxation times (τ_0, τ_1 and τ_2) and dipole moment (μ) of the samples at four different temperatures are mentioned in the above tables for all the three respective benches. From the tables it is clear that these relaxation times are decreasing with the increase in temperature. As we increase the temperature the values of average relaxation time (τ_0) decreases[16-17]. This is possibly due to increase in the molar volume[18].

The relaxation process of the molecules arise due to not only its intermolecular rotation but an intra-molecular rotation also contribute to it which shows that there is an additional intra molecular relaxation process exist to the overall relaxation process.

Table I, II and III also show that the value of τ_2 is higher than the average[18] relaxation time τ_0 which suggest the presence of intra-molecular bonding in the dilute solutions. The relaxation time depends upon the size and shape of the molecular entities in the solutions[19]. The values of relaxation time is minimum for X-band and maximum for C-band at a particular temperature. This shows that it depends on frequency of microwave used. It reveals that for the lower frequency relaxation time is higher whereas for the higher frequency relaxation time is lower for given samples.

The values of the dipole moments[20-21] of different samples are found to decrease with temperature which can be seen from the above tables. The decrease in the dipole moment may be due to the decrease in the size of dipole as the temperature increases[17]or with temperature the molecules tend to disorient due to increase in their thermal energy. The stretching of band moment and the change in the bond moment may be responsible for it. The values of dipole moment are independent of frequency as can be seen from the tables.

IV. Conclusion

The dielectric parameters of paracetamol in the dilute solution of carbon tetrachloride for five different mole fraction have been studied at four different temperatures using X-band, J-band and C-band microwave benches. From the results, it can be concluded that dielectric parameters and relaxation times are frequency and temperature dependent quantities whereas the dipole moment is a time dependent quantity.

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