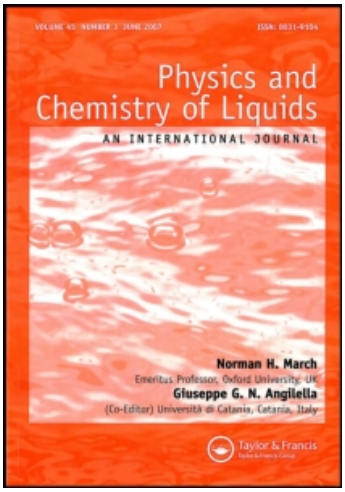

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ULTRASONIC STUDY ON 2-AMINOPYRIDINE IN SOLUTION IN BENZENE AT 309 K

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The ultrasonic velocity at 2 MHz, density, viscosity and refractive-index are measured at various concentrations in the solution of 2-aminopyridine in benzene at 309 K. Using the measured data, various useful parameters like adiabatic compressibility, Rao number, Wada's constant, Van der Waals constant, specific acoustic impedance, molar refraction, classical absorption, dielectric relaxation time, intermolecular free length and collision factor are computed. 2-Aminopyridine is found to be associative, showing strong molecular interactions with benzene.

KEY WORDS: Ultrasonic parameters, molecular interactions.

1 INTRODUCTION

The ultrasonic investigations due to simplicity of the technique reproducing accurate results, are being much widely used in the study of molecular behaviour in liquids[1–4]. Determination of various ultrasonic parameters such as ultrasonic velocity (C), adiabatic compressibility (β_a) Rao number (R_n), van der Waals constant (b) etc. leads to draw an idea regarding the molecular structure of liquids[5]. We have made ultrasonic study on a series of substituted compounds of pyridine with $-CH_3$ group, in various physical conditions and environments[6,7]. In this work we are now reporting a study on 2-aminopyridine, (a pyridine compound substituted with a $-NH_2$ group) being used in manufacture of drugs and pharmaceuticals, especially antihistaminic drugs. Though 2-aminopyridine is being studied currently[8], but its ultrasonic study has not been reported so far.

The compound being crystalline at room temperature (M.P. 58.1°C) has been studied in solution in benzene, at 309 K.

2 EXPERIMENTAL SECTION

The velocity at 2 MHz frequency has been measured using the interferometric method, measuring the wavelength (λ) at each concentration to an accuracy of

$\pm 0.3\%$. A single crystal variable path interferometer (Mittal's M-83 model) has been used. The density, viscosity and refractive-index were measured using a bicapillary pycnometer, Ostwald's viscometer and Abbe's refractometer respectively as described elsewhere[7]. The temperature during measurements was maintained to within ± 0.2 K by a water circulation thermostat (MLW U15C). The 2-aminopyridine 98% pure was procured from Ms. A. G. Fluka, Switzerland and benzene of analytical reagent grade, from E. Merck, India. From the measurement data various ultrasonic parameters are computed using the following relations[9]:

adiabatic compressibility—

$$\beta_a = (\rho C^2)^{-1} \tag{1}$$

Rao number—

$$R_n = (M/\rho)C^{1/3} \tag{2}$$

Wada's constant—

$$B_w = (M/\rho)\beta_a^{-1/7} \tag{3}$$

van der Waals constant—

$$b = (M/\rho)\left[1 - \frac{RT}{MC^2}\left\{\sqrt{1 + \frac{MC^2}{3RT}} - 1\right\}\right] \tag{4}$$

Specific acoustic impedance—

$$Z = \rho \cdot C \tag{5}$$

Molar refraction—

$$M_R = \left(\frac{M}{\rho}\right)\frac{n_D^2 - 1}{n_D^2 + 2} \tag{6}$$

Classical absorption—

$$\alpha_c = \frac{8\pi f^2 \eta}{3\rho C^3} \tag{7}$$

Dielectric relaxation time—

$$\tau_D = 3b\eta/4RT \tag{8}$$

Intermolecular free length—

$$L_f = \frac{2V}{Y}\left(1 - \frac{C}{C_\infty}\right) \tag{9}$$

Collision factor—

$$S = 4 \cdot \frac{C}{C_\infty} \cdot \frac{V}{b} \tag{10}$$

where C_∞ = 1600 M/S
Y = surface area per mole
R = Gas constant
T = absolute temperature
V = molar volume of the liquid
f = frequency of ultrasonics

In binary solutions the concentration dependence of the ultrasonic parameters is given by the relations

$$X = X_2 f_2 + X_1(1 - f_2) \tag{11}$$

where X = any parameter in solution
f_2 = mole fraction of solute

And suffix 1 is for solvent and 2 for the solute. Using the above relations the parameters have been evaluated at different concentrations of 2-aminopyridine in benzene.

3 DISCUSSION OF THE RESULTS

The measurement data of C, ρ, η and n_D at different solute concentrations and 309 K temperature are given in Table 1. The computed parameters from the measured data are given as β_a, R_n, B_w, b and Z in Table 2 and M_R, α_c/f^2, τ_D, L_f and S in Table 3. The concentration dependence of C and β_a are shown in Figure 1 and that of η and α_c/f^2 in Figure 2.

At our temperature of study 309 K, the solutions beyond 0.44 mole fraction concentration became turbid, so we kept our measurements limited up to $f_2 = 0.439$. From an observation of Figure 1 it is clearly seen that velocity increases non-linearly

Table 1 Measured data of C, ρ, η and n_D at various concentrations of 2-aminopyridine in benzene solution at 309 K.

Mole fraction f_2	*Ultrasonic velocity* C M/S	*Density* ρ gm/cm^3	*Shear index* η C_{poise}	*Refraction index* n_D
0.000	1248	0.857	0.5340	1.489
0.034	1263	0.861	0.5130	1.494
0.064	1269	0.866	0.5630	1.496
0.143	1289	0.886	0.6393	1.503
0.200	1307	0.904	0.7805	1.510
0.269	1329	0.917	0.9007	1.517
0.300	1343	0.931	0.9247	1.519
0.379	1369	0.945	1.0727	1.526
0.439	1376	1.246	1.6652	1.535

Table 2 Concentration dependence of β_a, R_n, B_w, Z and b in the solution of 2-aminopyridine in benzene at 309 K.

Mole Fraction f_2	$\beta_a \times 10^{11}$ $Cm^2/dy.$	R_n	B_w	$Z \times 10^{-4}$ $g\,S^{-1}\,cm^{-2}$	b $cm^3/mol.$
0.000	7.49	4555	2548	10.70	85.23
0.034	7.36	4636	2590	10.70	86.42
0.064	7.17	4595	2572	11.00	85.56
0.143	6.79	4585	2573	11.40	84.72
0.200	6.47	4564	2568	11.80	84.05
0.269	6.18	4589	2584	12.20	84.14
0.300	5.96	4562	2573	12.50	83.40
0.379	5.64	4589	2593	12.90	83.77
0.439	4.24	3528	2122	17.10	65.93

with the inclusion of 2-aminopyridine molecules in benzene, while due to closer packing of molecules in the solution adiabatic compressibility decreases. The changes in C and β_a are larger near 0.4 mol. fraction. These variations in C and β_a clearly indicate complex formations due to solute solvent interactions. From Table 1 density and velocity both are found increasing with f_2 thus the specific acoustic impedance is also increased as given in Table 2. From an observation of Tables 1 and 3 and Figure 2, it is found that viscosity increases non-linearly with concentration and similarly the classical absorption and dielectric relaxation time. The peaks in the absorption curve may be due to large intermolecular forces, exhibiting the associative nature of 2-aminopyridine molecule. It is also seen that absorption maxima occur at the compositions at which the shear viscosity also shows a maximum. This may be indicative of the formation of associated complex between solute and solvent molecules at those concentrations[10].

The molar refraction of 2-aminopyridine computed theoretically using the atomic bond lengths[11] was found to be 26.22, being a temperature independent quantity. The

Table 3 Concentration dependence of L_f, S, M_R, τ_D, α_c/f^2 in the solution of 2-aminopyridine in benzene at 309 K.

Mole fraction f_2	$L_f \times 10^8$ *cm.*	S	M_R	$\tau_D \times 10^{12}$ *sec.*	$\alpha_c/f^2 \times 10^{17}$ $S^2\,cm^{-1}$
0.000	1.277	3.33	26.31	13.3	8.4
0.034	0.278	3.38	26.60	12.9	7.9
0.064	0.273	3.39	26.71	14.1	8.4
0.143	0.256	3.45	26.83	15.8	10.0
0.200	0.242	3.50	26.90	19.1	10.9
0.269	0.223	3.26	27.21	22.1	11.0
0.300	0.212	3.34	27.04	22.5	10.8
0.379	0.190	3.49	27.33	26.2	11.7
0.439	0.183	3.56	21.79	32.0	13.5

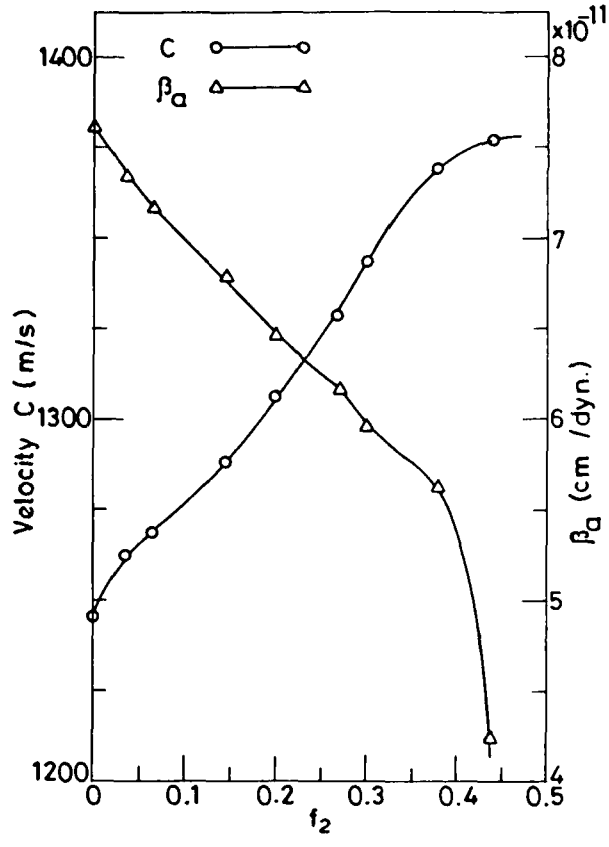

Figure 1 ○ ultrasonic velocity and △ adiabatic compressibility vs. concentration, in solution of 2-amino pyridine in benzene at 309 K.

experimental value of M_R for 2-aminopyridine, using Eq. (11) and M_R data from Table 3 was found to be 27.88, which is in near agreement with the theoretical value. The van der Waals b varies with mole fraction, as seen from Table 2, in conformity with M_R values, both being linearly related to each other[12]. The Rao number and Wada's constant both are found to vary non-linearly with f_2, as seen from Table 2. Both being molecular properties indicate the changing molecular interactions with changing solute and solvent ratio in the solution. From Table 3 it is observed that intermolecular free length decreases while Collision factor increases with f_2. The smaller values of L_f indicate closer packing of molecules in the solution exerting larger forces over each other which is reflected by increased values of molecular collision factor S up to 3.53, the maximum values of S can be 4.0.

From an overall behaviour of various ultrasonic parameters evaluated, it is concluded that 2-aminopyridine is a strong associative molecule, showing large interactions with benzene molecule.

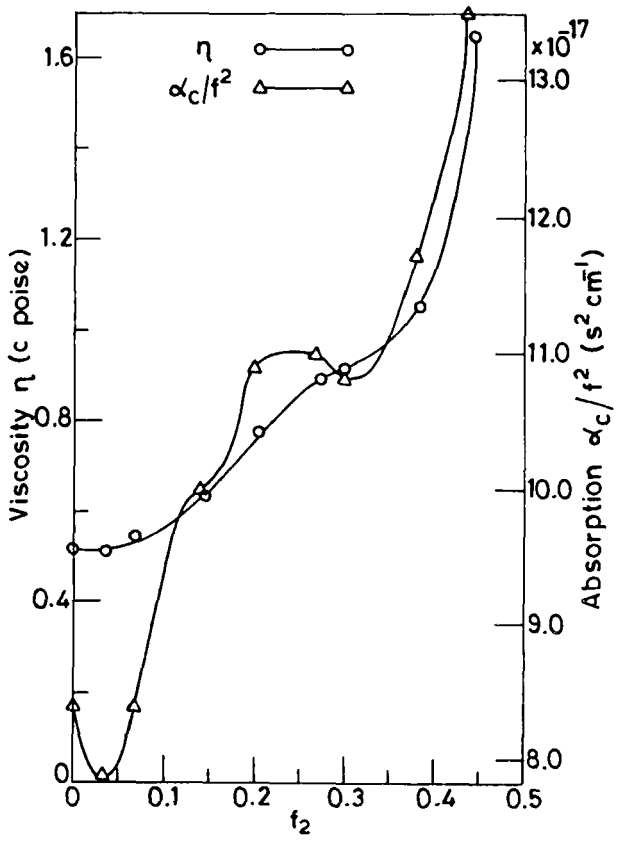

Figure 2 ○ viscosity and △ classical absorption vs. concentration in solution of 2-amino pyridine in benzene at 399 K.

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