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Research Article

INVESTIGATION OF ULTRASONIC VELOCITY AND RELATED ACOUSTICAL PARAMETERS OF N–(2-HYDROXYBENZYLIDENE)-3-SUBSTITUTED PYRIDINE-2-AMINE SCHIFF BASE DERIVATIVES IN ETHANOL-WATER MIXTURE AT 293, 297 AND 300 K

***Mrunal M. Mahajan and Pravin B. Raghuwanshi**

Department of Chemistry, BrijlalBiyani Science College, Amravati – 444602 (M.S.), India

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ABSTRACT

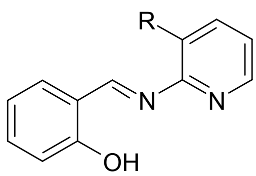
The density, viscosity and ultrasonic velocity of synthesized Schiff base N – (2-hydroxybenzylidene) pyridine-2-amine and its substituted derivatives have been studied in ethanol-water mixture at 293, 297 and 300 K over a wide range of concentration. From these experimental data, various acoustic parameters like apparent molar compressibility (ϕ_k), specific acoustic impedance (Z) and relative association (R_A) have been evaluated. The results are interpreted in terms of molecular interactions like solvent-solvent, solvent-solute and solute-solute interactions.

INTRODUCTION

Ultrasounds are sound waves with frequencies higher than the upper audible limit of human hearing. Ultrasound is used in many different fields. Ultrasonic devices are used to detect objects; measure distances and has imaging applications (Papadakis, 1999; economist.com., 2015). The measurement of ultrasonic speed along with other experimental data such as density and viscosity enables determination of some useful parameters, which are highly sensitive to molecular interactions (Jain *et al*, 1974; Wadekar, 2013; Jamankar *et al*, 2015; Burghate *et al*, 2016). Thus, ultrasonic velocity is helpful to interpreted solute-solvent, ion-solvent interaction in aqueous and non-aqueous medium.

Schiff bases are the heterocyclic compounds having high synthesis flexibility, varied coordinating ability and medicinal utility. Decent number of Schiff bases has been synthesized and reported for their bactericidal, fungicidal, antipyretic, antitumor, antitubercular and anticancer activity. Schiff bases create their own prominence in medicinal, pharmaceutical and agricultural science and hence it was thought interesting to investigate the reactions (Wahhenri Li *et al*, 2001; Raghuwanshi, 2012; Merchand, 1970). Number of researchers has investigated ultrasonic study of Schiff bases and reported about the variation in ultrasonic velocity with ion concentration, temperature and substituent. They also studied solute-solvent interaction, solvation number and other ultrasonic parameters (Deshmukh *et al*, 2010; Gangani *et al*, 2014; Premalatha *et al*, 2014).

Thus in the present study, efforts have been made to study the structural and molecular interactions of water molecules and organic solvent molecules with substituted Schiff bases, measured in terms of acoustic parameters like apparent molar compressibility (ϕ_k), specific acoustic impedance (Z) and relative association (R_A) in the suitable percentage of ethanol – water mixture at different temperatures.



Where,
R = -H, -OH, -NO_2, -CH_3

N - (2 - hydroxybenzylidene) pyridine - 2 - amine

Scheme I Structure of Schiff base derivative (A_1-A_4)

Corresponding author:* **Mrunal M. Mahajan
Department of Chemistry, BrijlalBiyani Science College, Amravati – 444602 (M.S.), India

1. N-(2'-hydroxybenzylidene) pyridine-2-amine (A_1)
2. N-(2'-hydroxybenzylidene)-3-hydroxy pyridine-2-amine (A_2)
3. N-(2'-hydroxybenzylidene)-3-nitropyridine-2-amine (A_3)
4. N-(2'-hydroxybenzylidene)-3-methylpyridine-2-amine (A_4)

MATERIALS AND METHODS

The chemicals used for synthesis were of L.R. grade. The ligands (A_1-A_4) were recrystallized before use. The solvent ethanol was purified using standard procedure. All the working solutions were freshly prepared from the deionized water to avoid any ionic contamination. The 0.01M solution of each ligand was prepared in different percentage (75%, 80%, 85%, 90%, 95% and 100%) of ethanol-water mixture. The density and the ultrasonic velocity measurements of the ligand solutions were done at 293, 297 and 300 K following the standard protocol.

Instrumentation

All the weighing in the present study was made on Citizen CY 104 one pan digital balance. The densities of the solution were determined by standardize capillary pyknometer having a bulb of volume of about 10 cm^3 and capillary having an internal diameter of 1 mm. A variable path ultrasonic interferometer from Mittal enterprises, New Delhi, Model MX-3 was used to measure the ultrasonic velocity in liquid mixtures and solutions, having the working frequency of 1 MHz with accuracy of ±0.03%.

RESULTS

From the experimental data of ultrasonic velocity, various acoustic parameters like apparent molar compressibility (ϕ_k), specific acoustic impedance (Z) and relative association (R_A) have been calculated. These acoustic parameters for ligands A_1, A_2, A_3 and A_4 were determined in varying percentage of ethanol-water and were studied at three different temperatures- 293, 297 and 300 K. The results are given in table 1 to12.

Table 1 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_1
Temp. = 293 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.87594	1.964	23836.33	1.2525	1.7203
80	0.86999	1.9758	19469.85	1.1864	1.7189
85	0.83164	1.9706	16924.60	1.0930	1.6388
90	0.81379	1.9676	-26767.04	0.8946	1.6012
95	0.8057	1.9682	25174.05	1.1716	1.5858
100	0.79878	1.793	33557.86	1.2085	1.4322

Table 2 Acoustic Parameters at different percentages of ethanol-water mixture.

System: Ligand – A_2
Temp. = 293 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8752	1.8564	27944.86	1.2752	1.6247
80	0.86408	1.987	19554.30	1.1762	1.7169
85	0.83553	1.964	16832.80	1.0994	1.6410
90	0.81868	1.8354	-21462.36	0.9211	1.5026
95	0.81696	1.7402	34911.56	1.2377	1.4217
100	0.80299	1.5446	49854.41	1.2768	1.2403

Table 3 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_3
Temp. = 293 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.87027	1.8082	30462.15	1.2792	1.5736
80	0.85605	1.7718	29132.44	1.2106	1.5167
85	0.83567	1.8128	23297.71	1.1293	1.5149
90	0.81657	1.777	-18238.24	0.9287	1.4510
95	0.81237	1.6096	43957.76	1.2632	1.3076
100	0.8043	1.6064	44759.82	1.2623	1.2920

Table 4 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_4
Temp. = 293 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6(kg m^{-2} sec^{-1})
75	0.87069	2.1086	19711.61	1.2159	1.8359
80	0.8606	2.1688	14324.44	1.1377	1.8665
85	0.83678	2.0002	15390.09	1.0943	1.6737
90	0.81761	2.1776	-34234.20	0.8689	1.7804
95	0.80842	2.239	15916.09	1.1261	1.8101
100	0.80132	1.9766	24655.88	1.1736	1.5839

Table 5 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_1
Temp. = 297 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8588	1.7266	35556.53	1.2818	1.4827
80	0.8491	1.5942	40236.82	1.2439	1.3537
85	0.8327	1.5950	36417.74	1.1744	1.3282
90	0.8201	1.5776	-5983.92	0.9704	1.2937
95	0.8169	1.6268	42071.24	1.2657	1.3289
100	0.8042	1.6248	43392.79	1.2574	1.3067

Table 6 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_2
Temp. = 297 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8577	1.6320	41128.83	1.3045	1.3998
80	0.8554	1.6216	37645.97	1.2459	1.3871
85	0.8204	1.3998	55609.23	1.2084	1.1484
90	0.8195	1.3788	12654.44	1.0142	1.1299
95	0.8104	1.3848	64909.98	1.3248	1.1222
100	0.8061	1.4454	58522.91	1.3105	1.1652

Table 7 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_3
Temp. = 297 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8520	1.7596	34579.08	1.2638	1.4992
80	0.8399	1.7482	32051.33	1.1931	1.4684
85	0.8346	1.6880	30119.77	1.1551	1.4089
90	0.8210	1.6332	-10093.29	0.9604	1.3409
95	0.8191	1.3310	69673.38	1.3569	1.0902
100	0.8036	1.4036	63495.29	1.3193	1.1280

Table 8 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_4
Temp. = 297 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8527	1.6430	41039.99	1.2940	1.4010
80	0.8457	1.6156	39236.85	1.2334	1.3663
85	0.8267	1.6292	34864.24	1.1576	1.3468
90	0.8258	1.6586	-12433.60	0.9610	1.3697
95	0.8121	1.6204	43200.84	1.2599	1.3159
100	0.8029	1.5826	46784.00	1.2663	1.2706

Table 9 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_1
Temp. = 300 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8586	2.1832	18499.74	1.1851	1.8744
80	0.8584	1.9574	21053.03	1.1742	1.6802
85	0.8539	2.0056	13780.54	1.1157	1.7126
90	0.8448	2.0510	-32475.07	0.9160	1.7327
95	0.8312	1.8466	27856.66	1.2347	1.5349
100	0.8178	1.8348	29214.06	1.2278	1.5005

Table 10 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_2
Temp. = 300 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8565	1.6432	40574.25	1.2997	1.4074
80	0.8445	1.8122	28350.09	1.1853	1.5303
85	0.8372	1.8152	23006.50	1.1309	1.5198
90	0.8215	1.7890	-19446.92	0.9321	1.4696
95	0.8181	1.7328	35193.02	1.2413	1.4177
100	0.8144	1.9858	23024.43	1.1910	1.6173

Table 11 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_3
Temp. = 300 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8554	1.6980	37492.57	1.2840	1.4525
80	0.8480	2.2114	14064.65	1.1138	1.8752
85	0.8450	1.8278	21642.14	1.1388	1.5445
90	0.8303	1.6116	-9857.49	0.9755	1.3380
95	0.8248	1.8062	30496.45	1.2342	1.4898
100	0.8155	1.3630	65852.29	1.3519	1.1115

Table 12 Acoustic Parameters at different percentages of ethanol-water mixture

System: Ligand – A_4
Temp. = 300 K

% Ethanol	d_s x 10^3 (kg m^{-3})	V x 10^3 (m sec^{-1})	$_k$ x 10^{-10} (m^3 mol^{-1} pa^{-1})	R_A	Z x 10^6 (kg m^{-2} sec^{-1})
75	0.8525	1.9300	27001.87	1.2261	1.6453
80	0.8362	1.8888	25728.99	1.1576	1.5795
85	0.8238	2.0260	15592.22	1.0728	1.6690
90	0.8195	1.9720	-27472.42	0.9002	1.6160
95	0.8124	1.8098	31684.13	1.2149	1.4703
100	0.8058	1.6374	42280.50	1.2565	1.3194

DISCUSSION

Apparent Molar Compressibility (ϕ_k)

The solute-solvent and solute-solute interactions in solutions can also be explained by acoustical parameter called apparent molar compressibility (ϕ_k). It explains the molecular interactions like structure making and structure breaking nature of solute. It is determined by measuring density and ultrasonic velocity and depends upon the molarity of solution and molecular weight of the solute. It is seen from table 1-12 and fig. 1-3 that ϕ_k values are negative for ligand solution in 90% ethanol showing interactions are insensitive to solvent. The positive values of ϕ_k show that electrostatic force in the vicinity of ion causes solvation of solute. It is well known that solute causing electrostriction leads to decrease in compressibility of solution which is reflected by negative values of ϕ_k (Dhanlakshmi *et al*, 1999)

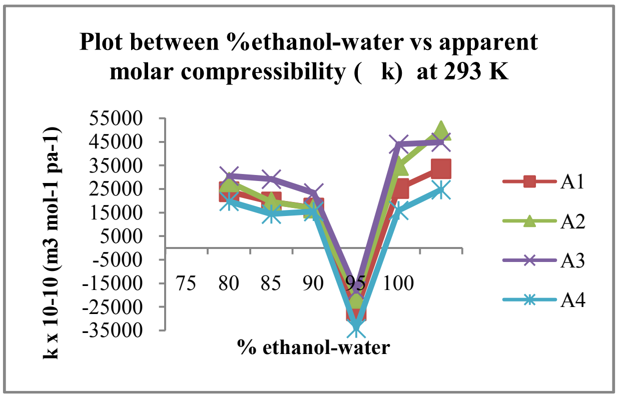


Fig 1

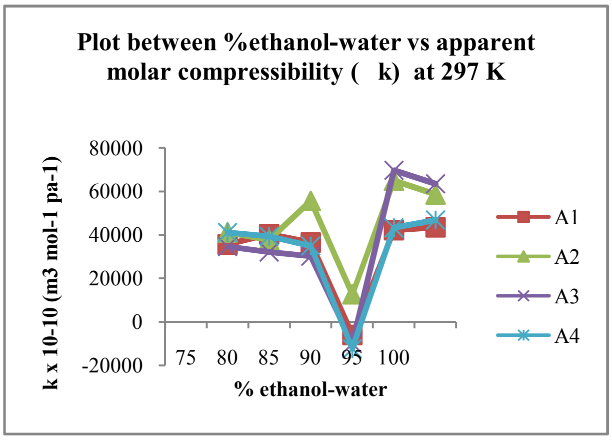


Fig 2

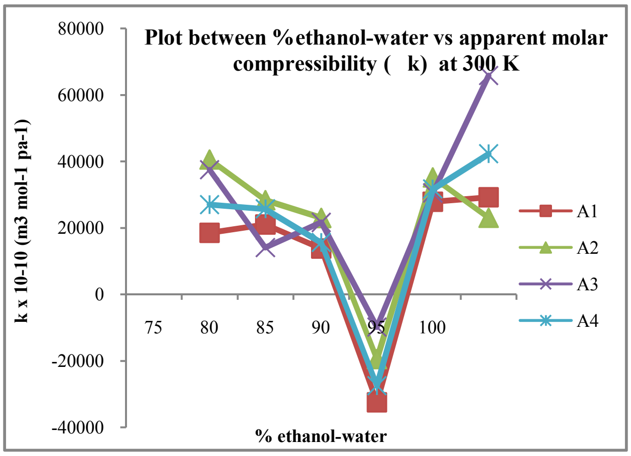


Fig. 3

Specific acoustic impedance (Z)

Specific acoustic impedance is defined as the impedance offered to the sound wave by the components of the mixture. Increasing trend of acoustic impedance supports the possibility of molecular interaction due to hydrogen bonding between Schiff base and ethanol. From table 1-12 and fig. 4-6, it can be seen that value of acoustic impedance varies with increase in concentrations. The trend observed as regards the variation in ultrasonic velocity with temperature. Moreover, lower values of Z in ethanol indicate hydrogen bonding (Mirikar *et al*, 2011).

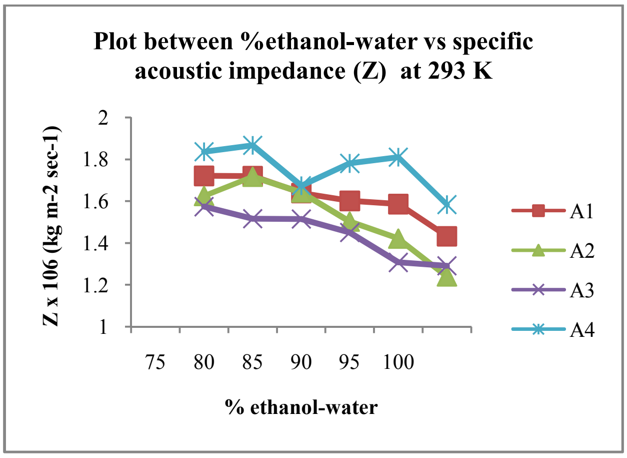


Fig 4

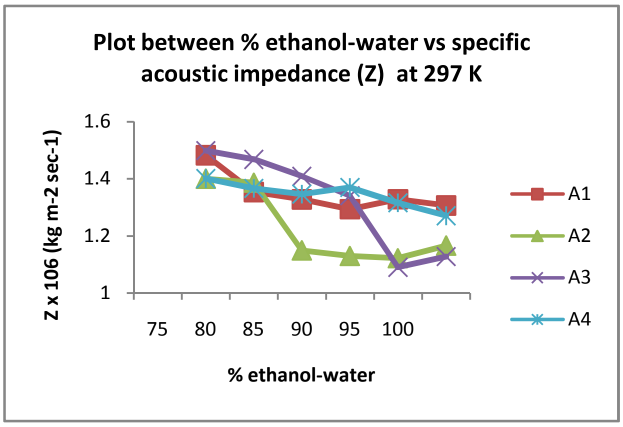


Fig 5

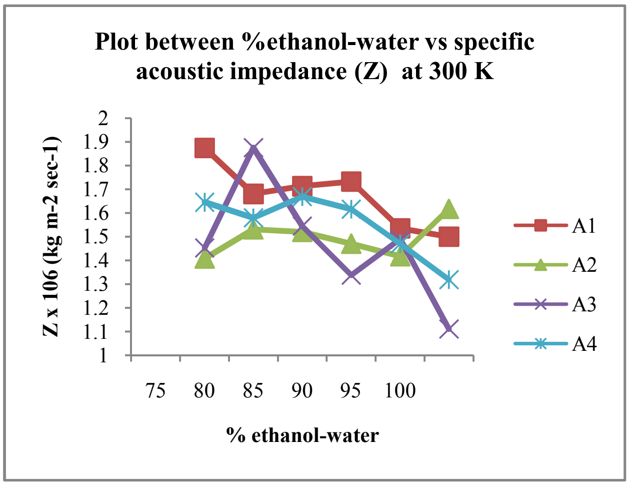


Fig 6

Relative association (R_A)

The alcohol molecules are well known to be associated. As soon as solute is added to alcohol, the probability of solute forming association with alcohol will be lesser. This is evident because the existing association in alcohol will render less probability of alcohol forming association with solute and hence has the lower R_A values (Erying *et al*, 1977). The data obtained is shown in table 1-12 and fig. 7-9.

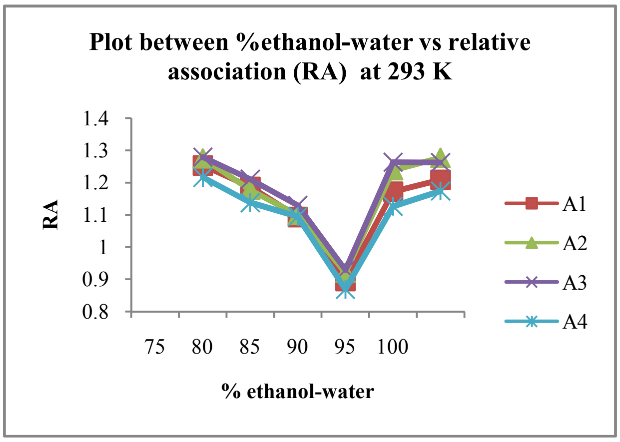


Fig 7

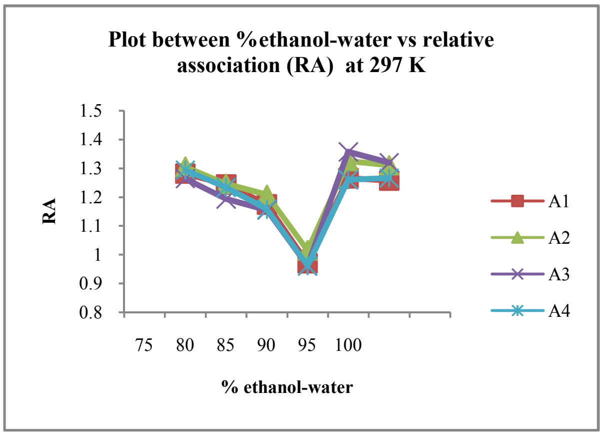


Fig 8

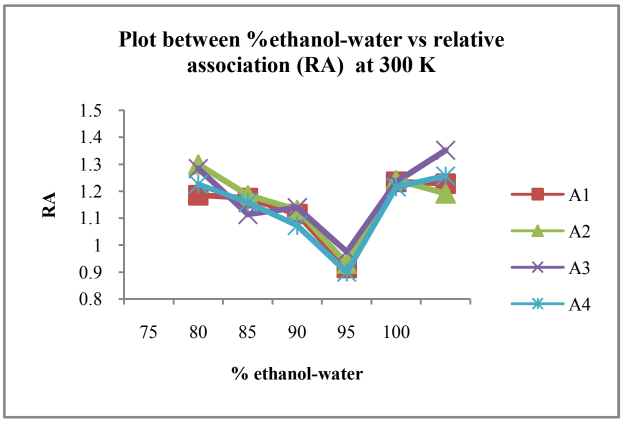


Fig 9

CONCLUSION

The acoustic parameters studied in the present work are in pure solvent ethanol, at 293, 297 and 300 K and interactions are interpreted. From this study, the structural and molecular interactions of water molecules and organic solvent molecules with substituted Schiff bases were studied and the properties which are directly or indirectly responsible for making and breaking the structure of the solvent were correlated. Thus measurement of ultrasonic velocity is the best tool to investigate structural interactions.

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