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Effect Of Temperature On Viscosity Of Pyrazolcarboxylic Acids Derivatives In 80% Acetone-Water Mixture At Different Temperature

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Abstract
Density and viscometric measurements of Substituted 1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid derivatives in binary mixture of 80% Acetone-water are performed. Jones-Dole empirical equation analyzed the obtained data . Thermodynamic parameters enthalpy change(ΔH) , entropy change (ΔS) and Gibbs free energy change(ΔG) is Determined. The interactions between solute-solute, solute-solvent and solvent–solvent interactions are studied.
Keywords: *Substituted 1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid derivatives, Jones-Dole empirical equation and Thermodynamic parameters.*

Introduction

Research on viscosity measurement yields crucial and significant data for pharmaceutical, and drug chemisty fields[1-2]. To predict solute–solvent interactions in the solution, volumetric and viscometric properties are helpful. solute–solvent interactions in the solution can be helpful in predicting the absorption of drugs and transport of drugs across the biological membranes. To understand drug action, the drug–water molecular interaction and their temperature dependence play an important role. The average free energy of activation of the pure solvent can be calculated at all the temperatures according to Eyring's model[3] Many industrial chemical processes or laboratory works need experimental data of viscosity at any given temperature and composition for binary liquid mixtures[4] viscosity and density of 1-((4-methoxy-3,5-dimethylpyridin-2-yl) methyl)-3-methylthiourea at controlled concentrations and varying temperatures is reported[5].Effect of temperature and chain length on the viscosity and surface tension of binary systems of N, N-dimethylformamide with 1-octanol, 1-nonanol and 1-decanol was studied[6]. Thermodynamic study of poly(vinyl pyrrolidone) in water/dimethyl sulfoxide solutions by viscometry is reported[7].Density and viscosity are studied at various temperature and composition of fatty acid and methyl esters from soyabean, castor and Jatrophacurcas oils[8].Influence of the temperature on the viscosity of magnetic ionic liquids was studied[9]. Viscosity of ammonium salts in water and ethanol-water systems was studied[10]. Gibbs free energy change, entropy change and enthalpy change in aqueous binary mixture of isomeric diols at different temperature was reported[11].Density and viscosity measurements for 4-aminobutyric acid in various composition in aqueous metformin hydrochloride at different temperature was reported[12]. Thermodynamic properties of sodium dodecyl sulfate in aqueous mixtures of methanol, ethanol, n-propanol and iso-propanol at different temperatures was reported[13].Density and viscosity of drug Paracetamol in various aqueous mixtures of ethanol have been determined[14].Densities and viscosities at different temperatures for aqueous binary mixtures of glycol-ethers at various concentrations range was studied[15].The metal complexes of Cu(II), Co(II), Ni(II) and Zn(II) with chromone Schiff base was reported[16].The literature survey shows the viscosity at different concentration of magnesium sulphate in mixture of formamide and ethylene glycol at room temperature was studied[17].The interaction of β-cyclodextrin with the amphiphilic drug promazine hydrochloride by using viscometric technique was reported[18]. Densities and viscosities of sodium ibuprofen in water at various temperature was studied[19].The volumetric and viscometric study of glycine at different temperature was done[20]. Viscometric study at different temperature of schiff-based resin of 2-Methoxy-1-Napthaldehyde was reported[21].

Viscosity for different molar concentration of L-Cerine, L-Proline and L-Histidine in dioxane-water mixture and evaluated the values of viscosity coefficients A and B of Jones-Dole equation have investigated[22]. The viscometric studies on the methyl cellulose-sodium dodecyl sulphate complex in aqueous solution was reported[23].Thermodynamic parameters of various drugs like substituted pyrazolin[24], substituted thiopyrimidine[25], nicotinic acid[26], amino acids[27], niacin[28], dodecyltrimethylammonium bromide[29], 1,3-diaryl carbamides[30], curcumin mixture[31] are reported.

In the present work, viscometric study of substituted1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid is carried out at different temperatures by using their solutions of different concentrations. Following substituted1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid are used.

1) **Ligand A (L_A)**= 1- phenyl-3-(4'- methyl) phenyl-1H- pyrazol-4-carboxylic acid
2) **Ligand B (L_B)**= 1- phenyl-3-(4'- bromo) phenyl-1H- pyrazol-4-carboxylic acid
3) **Ligand C (L_C)**= 1- phenyl-3-(4'- ethyl) phenyl-1H- pyrazol-4-carboxylic acid
4) **Ligand D (L_D)**= 1,3-diphenyl-1H- pyrazol-4-carboxylic acid

The solutions of ligands are prepared in the 70% solvent-Water at different temperature (*T*= 300K, 305K, 310K and 315K) and at different concentration.

Experimental

The ligands of which physical parameters are to be explored are synthesized by using reported protocol [32]. The A R grade solvent and freshly prepared doubly distilled water are used. The densities of pure solvent and solutions of various concentrations are measured at different temperatures using a specific gravity bottle. All the weights are taken on one pan digital balance (petit balance AD-50B) with an accuracy of ± 0.001 gm. Viscosities of the solutions are determined with the help of calibrated Ostwald viscometer (± 0.11% $Kgm^{-1}s^{-1}$). The flow time of solutions are measured by using digital clock (Racer Company) having an accuracy up to ±0.01Sec.

Results And Discussion

To calculate the relative and specific viscosity the following empirical equations are useful[33].

$$\eta_r = \frac{\eta}{\eta_o} = \frac{d_s \times t_s}{d_o \times t_o} \quad (1)$$

$$\eta_{sp} = (\eta_r - 1) \quad (2)$$

Where,

η_r	Relative viscosity
η_{sp}	Specific viscosity
d_s	Density of solution
η	Viscocity of solution
η_o	Viscosity of solvent
d_o	Density of solvent
t_o	Flow time for solvent
t_s	Flow time for solution

The viscosity data can be analyzed by Jones-Dole empirical equation[34].

$$\frac{(\eta_r - 1)}{\sqrt{C}} = \frac{\eta_{sp}}{\sqrt{C}} = A + B\sqrt{C} \quad (3)$$

Where,

A = Falkenhagen coefficient
B = Jones-Dole coefficient
C = Concentration of solutions

The Falkenhagen coefficient (*A*) reflects the solute-solute interactions while Jones-Dole coefficient (*B*) reflects the solute-solvent interactions.[1]

The thermodynamic parameters can be calculated by using following equations[35].

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$$\Delta G = -2.303\ R \times Slope \quad (4)$$

$$\log \frac{\eta_{r1}}{\eta_{r2}} = \left(\frac{\Delta H}{2.303}\right)\left[\frac{1}{T_1} - \frac{1}{T_2}\right] \quad (5)$$

$$\Delta S = \frac{(\Delta H - \Delta G)}{T} \quad (6)$$

In present study with increase in concentration, the relative viscosity and density of the compounds increases. The increase in viscosity with increase in concentration credited to the increase in the solute-solvent interactions. The viscosity and density data for different ligands at different concentration is given in table no.1

Table1: Densites(d) and viscosities(η_r) of substituted1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid derivatives of different concentration in 80% (Acetone+ water) solvent at 300K.

Conc. (C) mol/lit	L_A		L_B		L_C		L_D	
	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)
0.01	1.0616	2.3727	1.0623	2.4641	1.0421	2.3013	1.0717	2.3165
0.005	1.0612	2.3209	1.0715	2.3118	1.0319	2.1698	1.0683	2.2
0.0025	1.0589	2.2189	1.0586	2.2221	1.0418	2.0709	1.0691	2.1536
0.00125	1.0576	2.0966	1.0688	2.0985	1.0428	1.9756	1.0712	2.0610
0.000625	1.0572	1.8944	1.0692	1.8946	1.0387	1.8425	1.0663	1.8175

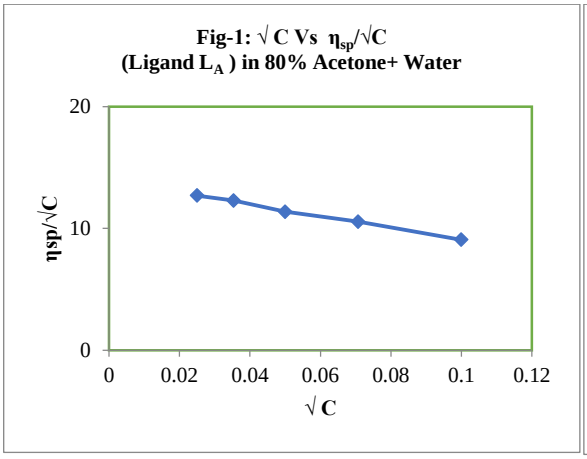
The graphs are plotted between √C versus $\eta_{sp}/\sqrt{C}$. The graphs for each system show the validity of Jones-Dole equation. The values of *A* and *B* have determined from the intercept and slope of √C versus $\eta_{sp}/\sqrt{C}$ respectively. The plots of √C versus $\eta_{sp}/\sqrt{C}$ for all four systems are shown in Fig. no. 1 to 5.

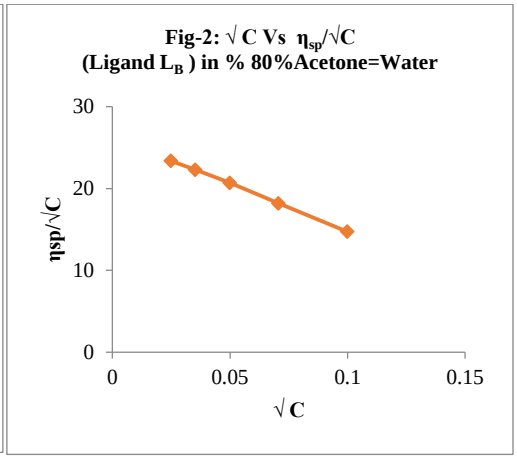
From the table no. 2 the *B*-coefficient is found to be negative for all the systems and it is a measure of disorder introduced by the solute into the solvent in all the systems. The Falkenhagen coefficient-*A* is positive in all the systems and this coefficient reflects strong solute-solute interaction.

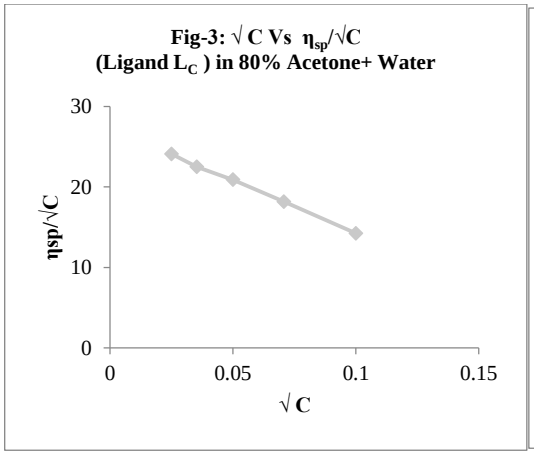
Viscometric study is performed for substituted1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid at following different temperatures; 300, 305, 310 and 315K. The experimental data of different ligands is presented in table no. 3.

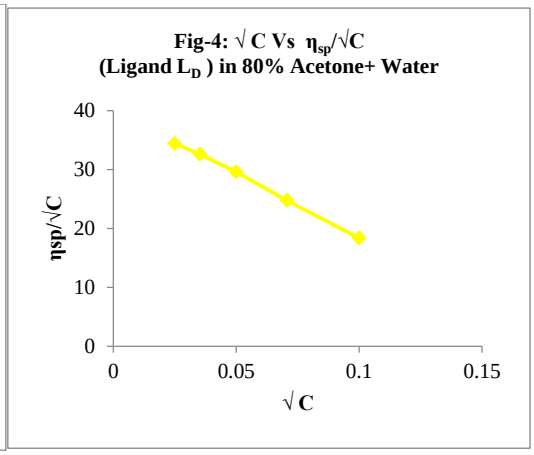
Table 2: *A* and *B* Coefficient values

Ligand + 80%Acetone-Water	*A* (lit$^{3/2}$ mol$^{-1/2}$)	*B* (lit mol^{-1})
L_A	13.786	-51.278
L_B	22.442	-110.09
L_C	27.310	-123.73
L_D	33.026	-213.56









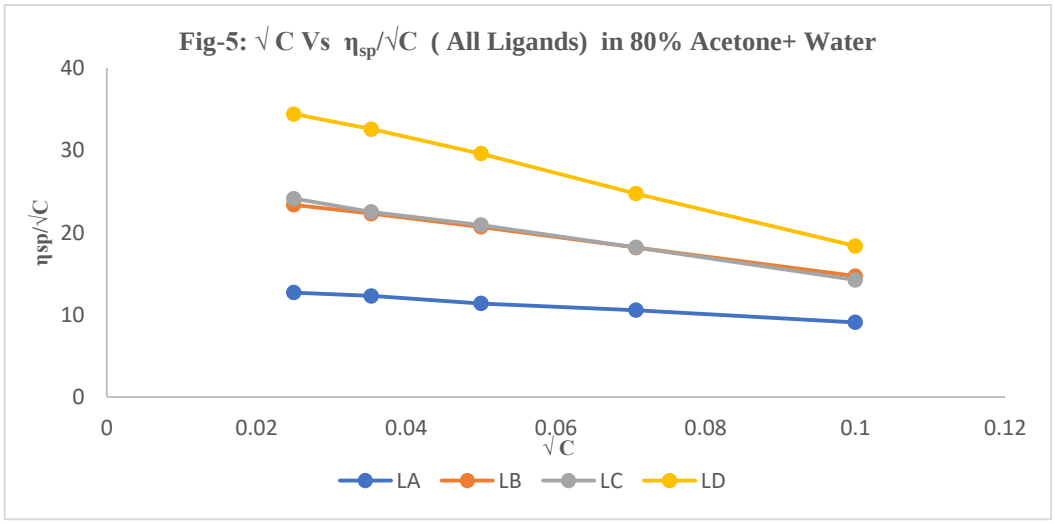


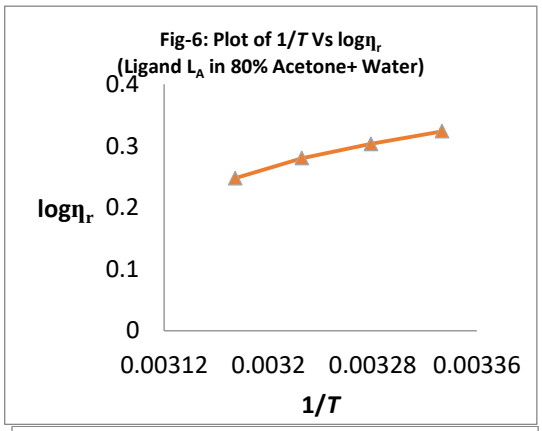
Table 3: Densites (d) and relative viscosities (η_r) of Substituted 1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid derivatives of 0.01M concentration in 80% (Acetone+ Water) solvent at different temperature (*T*= 300, 305, 310 and 315K).

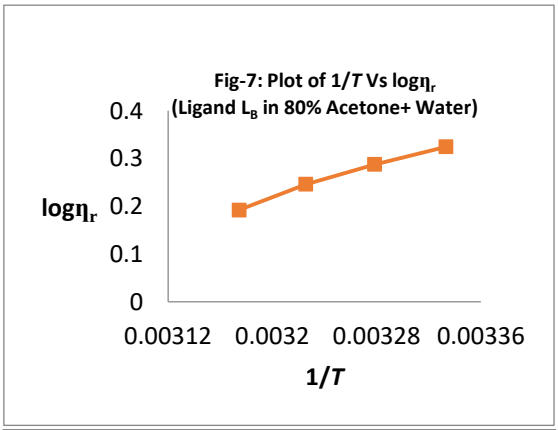
Temp. (K)	L_A		L_B		L_C		L_D	
	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)	Density (d) gm/cc	Relative Viscosity (η_r)
295	1.0611	2.1063	1.0722	2.3732	1.0431	1.0437	1.0698	2.3321
300	1.071	2.0104	1.0721	2.2231	1.1436	1.1428	1.0621	2.2037
305	1.0619	1.9051	1.0705	2.0832	1.2035	1.2027	1.0521	2.0259
310	1.0615	1.7678	1.0692	1.9929	1.1971	1.1965	1.0407	1.8801

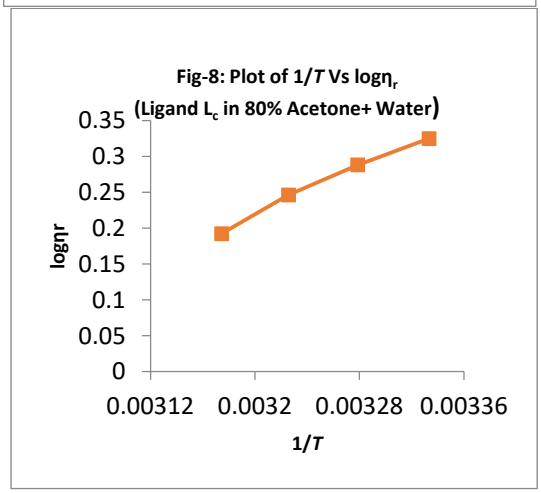
The thermodynamic parameter Gibbs free energy change (ΔG), enthalpy change (ΔH) and entropy change (ΔS) are studied for substituted 1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid at different temperatures. All these thermodynamic parameters are calculated by plotting graphs between 1/T versus logη_r. These are shown in the fig. no. 6 to 10. These thermodynamic parameters for solutions of different ligands at various concentrations are presented in table no. 4. The values of ΔG and ΔH are found to be negative. The negative values of ΔG and ΔH indicate the reactions are spontaneous and exothermic respectively. The negative value of ΔS indicates that there is an association of solvent molecules around the ligand.

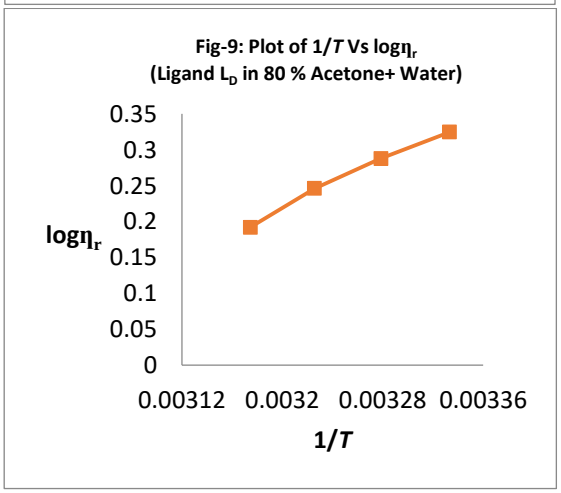
Table 4: Values of thermodynamic parameters for temperature difference (300 to 315K)

Ligands	ΔG (J mol^{-1})	ΔH (J mol^{-1})	ΔS (J mol^{-1} K^{-1})
L_A	-1289.2	-4289.1	-97.79
L_B	-1035.5	-3341.3	-74.41
L_C	-1069.5	-3393.7	-73.36
L_D	-1811.2	1505.2	-45.17









Conclusions

In the present work, viscometric study is performed at different temperatures for substituted 1-phenyl-3-aryl-1H-pyrazol-4-carboxylic acid. It is found that the density and relative viscosity increases as the concentration increases. And it is attributed to the concentration increases the increase in the solute-solvent interaction. *B*-coefficient is found to be negative for all the systems and it measures the disorder introduced by solute into the solvent. The Falkenhagen coefficient-*A* is positive in all the systems and this coefficient reflects strong solute-solute interaction in systems. The negative values of ΔG and ΔH indicates the reactions are spontaneous and exothermic. The negative values of ΔS indicate that there is an association of solvent molecules around the ligand.

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