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Ultrasonic Studies on Molecular Interaction of Mesitylene With Alkanols in Cyclohexane Solution at 303K

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ABSTRACT *Ultrasonic velocity(U), Density(ρ) and Viscosity ( ) have been measured for mixtures of alkanols namely 1-Propanol, 2-Propanol, 1-Butanol and 1-Pentanol with Mesitylene in Cyclohexane at 303K. The experimental data was used to calculate various acoustical parameters like adiabatic compressibility( ), Free Length(Lf), Free Volume (Vf), Internal Pressure (πi), Acoustical Impedance (Z), Absorption Coefficient, Avilable Volume, Cohesive Energy, Lenard Jones potential, Free energy of Activation, Molecular interaction parameter and Formation Constant. These parameters are used to discuss the Molecular interactions in the Mixtures.*

KEYWORDS : Ultrasonic Velocity, Acoustic parameters, Dipolar and dispersive interactions.

INTRODUCTION:

The Ultrasonic studies in liquids are great use in understanding the nature and strength of Molecular Interaction. A large number of studies have been made on the intermolecular interaction in liquid system by various methods like Ultraviolet, Dielectric constant, Infrared, Raman Effect, Nuclear magnetic resonance and Ultrasonic method. In recent years Ultrasonic method has become a powerful tool in providing information regarding the physico-chemical properties of liquid systems [1-4]. The method of studying in Molecular Interaction from the knowledge of variation of Acoustic parameters values with change in concentration. The increase (or) decrease in Ultrasonic velocity have been used in understanding the nature of molecular interactions in the pure liquids of Ternary Mixtures. The variation of Ultrasonic Velocity and Related parameters show much light upon structural changes associated with the liquid upon structural changes associated with the liquid mixtures having weakly interacting components as well as strongly interacting components. The study of liquid mixtures containing of polar and non-polar components find solutions that generally do not behave ideally.

In the present study Ultrasonic velocity, Density and Viscosity were measured experimentally for the ternary systems namely Mesitylene and cyclohexane with 1-propanol, 2-Propanol, 1-Butanol and 1-Pentanol at 303K and predicted the possible Molecular Interaction between the unlike molecules. The significance of acoustic studies in mixtures have been used for understanding the intermolecular interactions by many researchers for interpreting different type of interactions like dipole – dipole [5-8] and dipole – induced dipole [9-13]. The presence of –CH_3 group in Mesitylene implies that dipole-induced interaction is weaker in hydrocarbons owing to its decrease polarisabilities with increase of –CH_3 group [14-21].

Materials and Methods:

The liquid mixtures of various concentration in were prepared by taking AR grade chemicals, which were purified by standard methods.

The Ultrasonic velocity in liquid mixtures have been measures using an Ultrasonic Interferometer (Mittal type: Model: F81) working at frequency 2MHz with an overall accuracy of ± 0.1 ms^{-1}. The density and viscosity are measure using a Pycknometer and an Ostwald's Viscometer with an accuracy of ± 0.1 kg m^{-3} and ±0.0001 m Nm^{-2}s, respectively. All the precautions were taken to minimize the possible experimental error. The set-up is checked for standard liquids.

The expression used to determine the Ultrasonic velocity is U = f λ ms^{-1}----------(1)

The densities of the mixture were measured using the formula $ρ_2$ = $(w_2/w_1)ρ_1$----------(2)

Where w_1 = weight of distilled water, w_2 = weight of experimental liquid, $ρ_1$ = Density of water, $ρ_2$ = Density of experimental liquid.

The viscosity was determined using the relation,

$Π_2 = Π_1 (t_2/t_1) (ρ_2/ρ_1)$ ----------------(3)

Where $Π_1$ = viscosity of water, $Π_2$ = Viscosity of mixture, $ρ_1$ = Density of water, $ρ_2$ = Density of mixtures, t_1 = Time of flow of water, t_2 = Time of flow of mixture.

Using the measured data, the following acoustical parameters can be calculated.

Adiabatic compressibility ( )

K == $(1/U^2ρ)$ $kg^{-1}ms^{-2}$ ------------------(4)

Free Length(L_f)

L_f == $(K/Uρ^{1/2})$ m ------------------------(5)

Where, K is Jacobson's constant. This constant is a temperature dependent parameter whose value at any temperature (T) is given by (93.875 + 0.345T) X 10^8.

Acoustic Impedance (Z)

Z == U ρ kg $m^{-2}s^{-1}$

Free Volume (V_f)

V_f == $(M_{eff} U/kΠ)^{3/2}$ m^3

Where, M_{eff} is the effective molecular weight (M_{eff} = $Σ m_i x_i$, in which m_i and x_i are the molecular weight and the mole fraction of the individual constituents respectively and k is a temperature independent constant equal to 4.28 x 10^9 for all liquids.

Internal pressure ($π_i$)

On the basis of statistical thermodynamics. Suryanarayana derived an expression for the determination of internal pressure through use of concept of free volume

$Π_i$ == bRT $(kΠ/U)^{1/2}$ $(ρ^{2/3}/M_{eff})^{7/6}$

Where

T = is the absolute temperature

P = is the density and R is the gas constant.

M_{eff} = is the effective molecular weight.

Absorption Coefficient (α/f^2):

It can be calculated from the viscosity using the relation,

$\alpha/f^2 = 8\pi^2 \eta/3\rho U^3$

Viscous Relaxation Time (τ)

It is calculated using the relation,

$\tau = 4\eta/3\rho U^2$

Available Volume (V_a):

It can be calculated from Schaff's relation

$V_a = V_m (1-U/U_a)$ m^3

Where V_m is the molar volume and $U_a = 1600$ ms^{-1}.

Cohesive Energy (CE):

It is usually given as a product of internal pressure (π_i) and molar volume(V_m).

$CE = \pi_i V_m$ kJ mol^{-1}

Free energy of Activation (ΔG^*):

The free energy of activation (ΔG^*) is calculated by

$(\Delta G^*) = -k_B T \ln (h/\tau k_B T)$ kJmol^{-1}

Where,

K_B = Boltzman's constant (1.3806×10^{-23} Jk^{-1})

T = is the temperature

h = is the Planck's constant (6.626×10^{-34} Js)

Lenard Jones Potential:

The Lenard Jones potential exponent is given by

$LJP = 6V_m/V_a$

V_m = the molar volume V_a = the available volume

Formation constant:

To calculate the formation constant values of the charge transfer complexes, appreciable to weak complexes and in very dilute solutions. The stability constant is calculated using the relation

$K = Y/(b-y)^2$ dm^3 mol^{-1}
Where, $Y = (a-k^{1/2}b)/k---k^{1/2}$);   k = x/y
X = difference between U_{cal} and U_{obs} at lower concentration 'a',
Y= difference between U_{cal} and U_{obs} at higher concentration 'b' and
U_{cal} = the ultrasonic velocity of the mixture.

This equation can be used to calculate stability constant values for different combination concentration 'a' and 'b'.

Result and Discussion:
The Measured Ultrasonic Velocity, Density and Viscosity at various equimolar concentration represented in Table 1,2 and 3. The variation of Ultrasonic velocity depends upon the increase (or) decrease of intermolecular free length after mixing the components. On the basis of a Model for propagation proposed by Kinaid and Eyring (1938), ultrasonic velocity should increase if the intermolecular free length decreases as a result of mixing of components and vice versa. This facts

is observed in the present investigation. The Dispersive type of interaction may occur between the alkanol and cyclohexane due to the non-polar nature of cyclohexane and its intertness towards electron donors. The addition of alkanols with the mixture leads to a compact structure due to the presence of dipolar (or) dispersive type interactions. This contributes to a decrease in free length and hence compressibility. The decrease in free length due to the close appearance of the molecules. The increase in free length is due to the loose packing of molecular inside the shield, which may be brought about by weaking of molecular interactions. Though, the interactions between the Aromatic derivatives set-up an interaction between π-electron could and the hydroxyl group (Stokes and Robinson, 1982). Though the interaction is of minor intensity compared with hydrogen bonding but they can lead to the formation of intermolecular complexes (Prigogine, 1957).Finally the contribution due to structural effects which may be taken into account which may result in decrease of adiabatic compressibility (β) and intermolecular free length.

A decrease in free volume and an increase in internal pressure to the increase in magnitude of interaction. As the dipole moment of cyclohexane is zero, the decrease in free volume shows that the clustering is not due to H-bonding but may be due to dispersive interactions. It is primarily due to the formation of spherical cage like structures owing to the closer packing of the molecules. As the number of –CH$_3$ group increases, the electron donating tendency increases in hydrocarbons, but at the same time there may be steric repulsion which hinder proper orientation. The increase of –CH$_3$ group in Mesitylene implies that dipole-induced dipole interactions is weaker in higher hydrocarbons (Mesitylene) owing to its decreases polarisabilities with the increase of –CH$_3$ groups. The calculated values of Adiabatic complressibility, Free Length, Free volume and Internal pressure are represented by table 4,5 and 6.

Cohessive Energy (CE) values of four systems are tabulated in table . The Cohessive Energy increases with increase in concentration for the four system of donor – acceptor systems. Cohessive Energy is a measure of attraction between molecules of the components. The increase in Cohessive Energy values with increase in concentration shows stronger molecular interactions are present in these system. Extent of complexation with increase in concentration for four system is also shown by increase in Lenard Jones potential with increase in concentration. Lenard Jones potential values are computed and presented in Table8.

Molecular Interaction parameter (χ_u) values are presented in Table   for these systems. Molecular Interaction parameter shows the extent of deviation from ideal behavior which may be due to charge transfer complex formation of four system exhibit negative deviation from ideal behavior shows complexation is concentration dependent. The values of free energy of Activation (ΔG^*) and Relaxation time (τ) for these system are presented in table7 and 9.

The values of free energy of Activation (ΔG^*) and Relaxation time (τ) suggest the charge-Transfer complex formed at various concentrations.

The formation constant value (K) for the four system are presented in Table10. The mean value of formation constant values are presented in Table 11.

The formation constant (K) increases with increases in polarizability of donor molecules. Therefore, increase in polarisability of donor increases the ease of complexation. But in the present study involves the dipole – induced dipole interactions is weaker in hydrocarbons (Mesitylene) owing to its decreases polarisabilities with the increase of –CH$_3$ groups.

Conclusion:
The result shows that Mesitylene forms donor – Acceptor complex with alcohols. The formation constant of these complexes indicates the presence of Electron releasing group in acceptor molecules. Hence, the formation constant is higher for Mesitylene & 2-Propanol than that , Mesitylene & 1-Propanol complex. Due to the steric effect and symmetrical nature of electron donors the formation constant value is low for Mesitylene & 1-Butanol and 1-Pentanol.

Table 1: Ultrasonic velocity (ms^{-1}) values of Mesitylene with Alakanol in Cyclohexane at 303K

Equimolar concentration	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10^{-3}	1192.4	1191.5	1179.0	1188.0
2X10^{-3}	1224.1	1221.7	1185.8	1180.0
3x10^{-3}	1191.0	1188.3	1183.0	1182.7
4X10^{-3}	1185.9	1184.1	1184.9	1184.6
5X10^{-3}	1170.5	1222.1	1183.3	1185.0
6X10^{-3}	1191.3	1184.6	1188.8	1186.4
7X10^{-3}	1187.2	1185.2	1183.9	1183.9
8X10^{-3}	1191.2	1214.6	1190.3	1182.6
9X10^{-3}	1186.2	1224.1	1186.5	1180.8
10X10^{-3}	1185.9	1220.2	1186.8	1179.8

Table:2 Density (Kg/m³) values of Mesitylene with Alkanol in cyclohexane at 303K

Equimolar concentration	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10^{-3}	728.3	733.3	726.4	726.3
2X10^{-3}	730.4	724.8	728.8	727.8
3X10^{-3}	726.8	725.4	726.7	728.9
4X10^{-3}	727.6	724.0	726.5	732.5
5X10^{-3}	729.6	726.5	724.4	725.2
6X10^{-3}	727.6	725.0	731.5	728.8
7X10^{-3}	726.4	726.4	725.5	727.0
8X10^{-3}	724.4	725.4	728.3	728.1
9X10^{-3}	731.3	726.2	729.3	729.2
10X10^{-3}	730.5	724.1	726.7	732.0

Table:3 Viscosity (10^{-4} Nsm^{-2}) values of Mesitylene with Alkanol in cyclohexane at 303K

Equimolar concentration	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10^{-3}	7.096	7.029	6.963	6.962
2X10^{-3}	7.002	7.176	7.101	6.977
3X10^{-3}	7.195	7.068	6.966	6.873
4X10^{-3}	7.432	6.940	6.964	7.022
5X10^{-3}	7.109	6.850	7.058	7.066
6X10^{-3}	7.089	7.064	7.012	7.101
7X10^{-3}	7.077	6.849	7.069	7.083
8X10^{-3}	6.944	6.840	7.096	7.208
9X10^{-3}	7.125	6.961	6.876	7.334
10X10^{-3}	7.232	7.055	6.966	7.132

Table:4 Adiabatic Compressibility(10^{-10} kg^{-1}ms²),& Free Volume (10^{-7}m³)) values of Mesitylene with alkanol in cyclohexane at 303K

Equimolar concentration	Adiabatic compressibility (10^{-10} kg^{-1}ms²)				Free volume V_f/10^{-7} m³			
	1-propanol	2-Propnol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10^{-3}	9.66	9.61	9.90	9.76	2.531	2.564	2.560	2.590
2X10^{-3}	9.14	9.24	9.76	9.87	2.016	1.937	1.882	1.918
3X10^{-3}	9.70	9.76	9.83	9.81	1.857	1.901	1.930	1.968
4X10^{-3}	9.77	9.85	9.80	9.73	1.758	1.943	1.935	1.911
5X10^{-3}	10.00	9.22	9.86	9.82	1.842	2.078	1.893	1.894
6X10^{-3}	9.68	9.83	9.67	9.75	1.899	1.894	1.925	1.883
7X10^{-3}	9.77	9.80	9.83	9.81	1.894	1.985	1.890	1.884
8X10^{-3}	9.73	9.35	9.69	9.82	1.959	2.063	1.894	1.832
9X10^{-3}	9.72	9.19	9.74	9.84	1.873	2.033	1.976	1.782
10X10^{-3}	9.73	9.28	9.77	9.81	1.831	1.983	1.939	1.855

Table:5 Internal pressure ($\pi_i$atm) & Acoustic Impedance Z/10^{5}kgm^{-2}S^{-1} Values of Mesitylene with alkanols in cyclohexane at 303K

Equimolar concentration	Internal pressure (π_i atm)				Acoustic ImpedanceZ/10^{5}kgm^{-2}s^{-2}			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10^{-3}	2927.45	2928.10	2911.26	2899.80	8.68	8.74	8.56	8.63
2X10^{-3}	3595.04	3624.39	3673.08	3646.37	8.94	8.85	8.64	8.59
3X10^{-3}	3682.68	3649.17	3635.40	3618.59	8.66	8.62	8.60	8.62
4X10^{-3}	3753.46	3617.95	3631.31	3666.70	8.63	8.57	8.61	8.68
5X10^{-3}	3701.78	3546.21	3651.10	3653.09	8.54	8.88	8.57	8.59
6X10^{-3}	3657.60	3652.55	3654.50	3672.17	8.67	8.59	8.70	8.65
7X10^{-3}	3656.86	3600.42	3656.62	3665.45	8.62	8.61	8.59	8.61
8X10^{-3}	3609.54	3550.95	3663.23	3703.44	8.63	8.81	8.67	8.61
9X10^{-3}	3687.32	3571.08	3615.30	3742.23	8.67	8.89	8.65	8.61
10X10^{-3}	3712.59	3593.87	3629.67	3701.46	8.66	8.84	8.62	8.64

Table:6 Free Length (L$_f$ pm) & Absorption Coefficient α/f² 10⁻¹⁴Npm⁻¹s² for Mesitylene with alkanols in Cyclohexane at 303K

Equimolar concentration	Free Length (L$_f$)				Absorption Coefficient α/f² 10^{-14} Npm⁻¹s²			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10⁻³	6.17	6.15	6.25	6.20	1.51	1.49	1.54	1.50
2X10⁻³	5.95	5.98	6.15	6.18	1.37	1.43	1.54	1.53
3X10⁻³	6.13	6.15	6.17	6.16	1.54	1.53	1.52	1.50
4X10⁻³	6.15	6.17	6.16	6.14	1.61	1.52	1.52	1.52
5X10⁻³	6.22	5.97	6.18	6.16	1.60	1.36	1.55	1.54
6X10⁻³	6.12	6.17	6.12	6.14	1.52	1.54	1.50	1.53
7X10⁻³	6.15	6.16	6.17	6.16	1.53	1.49	1.54	1.54
8X10⁻³	6.14	6.01	6.12	6.16	1.49	1.38	1.52	1.57
9X10⁻³	6.13	5.96	6.15	6.16	1.56	1.41	1.51	1.56
10X10⁻³	6.14	5.99	6.15	6.16	1.56	1.41	1.51	1.56

Table:7 Viscous Relaxation time τ/10⁻¹³ S & Available Volume V$_a$/10⁻⁵ m³ for Mesitylene with alkanols in Cyclohexane at 303K

Equimolar concentration	Viscous Relaxation time τ/10^{-13} s				Available Volume V$_a$/10^{-5} m³			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10⁻³	9.137	9.003	9.194	9.056	3.595	3.548	3.604	3.605
2X10⁻³	8.530	8.844	9.239	9.179	2.886	2.746	2.893	2.897
3X10⁻³	9.307	9.200	9.133	8.987	2.750	2.985	2.750	2.742
4X10⁻³	9.684	9.116	9.104	9.108	2.560	3.022	2.564	2.543
5X10⁻³	9.482	8.418	9.278	9.251	2.647	2.736	2.666	2.663
6X10⁻³	9.154	9.257	9.044	9.229	2.956	3.013	2.940	2.951
7X10⁻³	9.217	8.950	9.268	9.268	2.421	3.004	2.424	2.419
8X10⁻³	9.007	8.522	9.169	9.438	2.669	2.795	2.655	2.656
9X10⁻³	9.233	8.530	8.930	9.618	2.650	2.723	2.657	2.657
10X10⁻³	9.386	8.726	9.074	9.334	2.820	2.759	2.834	2.814

Table:8 Cohesive Energy CE KJ/mole & Lenard Jones Potential for Mesitylene and alkanols in Cyclohexane at 303K

Equimolar concentration	Cohesive Energy CE KJ/mole				Lenard Jones Potential			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10⁻³	70.566	70.085	70.751	70.761	10.355	10.502	10.355	10.355
2X10⁻³	57.871	58.318	57.998	58.077	10.952	12.375	10.952	10.952
3X10⁻³	58.499	58.612	58.507	58.330	12.263	10.319	12.263	12.263
4X10⁻³	58.393	58.683	58.481	58.002	14.113	10.081	14.113	14.113
5X10⁻³	57.661	57.907	58.075	58.011	13.149	12.402	13.419	13.419
6X10⁻³	58.185	58.394	57.875	58.089	10.476	10.112	10.476	10.476
7X10⁻³	58.090	58.090	58.162	58.042	15.708	10.144	15.708	15.708
8X10⁻³	58.533	58.452	58.219	58.235	13.115	11.907	13.115	13.115
9X10⁻³	57.980	58.387	58.139	58.147	13.059	12.537	13.059	13.059

Table:9 Free energy of Activation ΔG* KJ/mole & Molecular Interaction parameter χu/10⁻² for Mesitylene and alkanol in Cyclohexane at 303K

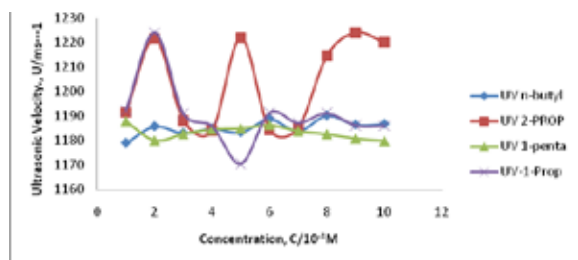| Equimolar concentration	Free Energy of Activation ΔG*KJ/mole				Molecular Interaction parameter χu/10^{-2}			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1X10⁻³	4.840	4.799	4.857	4.816	-6.326	-6.463	-8.414	-7.017
2X10⁻³	4.650	4.750	4.871	4.853	-1.283	-1.667	-7.368	-8.267
3X10⁻³	4.891	4.859	4.839	4.795	-6.553	-6.964	-7.807	-7.849
4X10⁻³	5.001	4.834	4.830	4.831	-7.343	-7.626	-7.508	-7.555
5X10⁻³	4.943	4.614	4.883	4.874	-9.741	-1.602	-7.759	-7.494
6X10⁻³	4.845	4.876	4.812	4.868	-6.505	-7.538	-6.898	-7.277
7X10⁻³	4.864	4.783	4.880	4.880	-7.145	-7.450	-7.662	-7.669
8X10⁻³	4.801	4.648	4.850	4.930	-6.519	-2.808	-6.662	-7.874
9X10⁻³	4.869	4.650	4.777	4.982	-7.309	-1.278	-7.265	-8.156
10X10⁻³	4.194	4.713	4.821	4.899	-7.347	-1.909	-7.216	-8.314

Table:10 Formation Constant (K/m^{-1}) for Mesitylene with alkanol in Cyclohexane at 303K

Equimolar Concentration	Formation Constant			
	1-Propanol	2-Propanol	1-Butanol	1-Pentanol
1×10^{-3}	88.9	88.3	80.7	85.9
2×10^{-3}	71.3	66.7	40.5	38.7
3×10^{-3}	47.6	46.8	45.2	45.1
4×10^{-3}	37.3	36.8	37.0	36.9
5×10^{-3}	28.7	41.5	30.8	31.1
6×10^{-3}	28.9	27.8	28.4	28.1
7×10^{-3}	24.9	24.6	24.5	24.5
8×10^{-3}	23.4	27.0	23.2	22.3
9×10^{-3}	20.8	20.2	20.8	20.2

Table:11

System	Mean K Value
Mesitylene & 1-Propanol	41.31
Mesitylene & 2-Propanol	42.18
Mesitylene & 1-Butanol	36.78
Mesitylene & 1-Pentanol	36.97

Figure 1. Plot of Ultrasonic velocity Vs Concentration
Solvent: Cyclohexane

Figure 2. Plot of Adiabatic compressibility Vs Concentration
Solvent: Cyclohexane
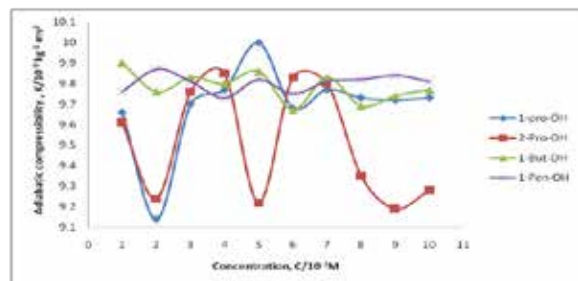
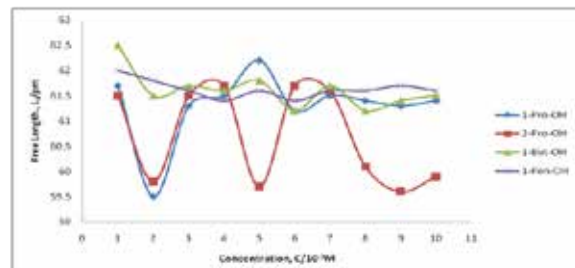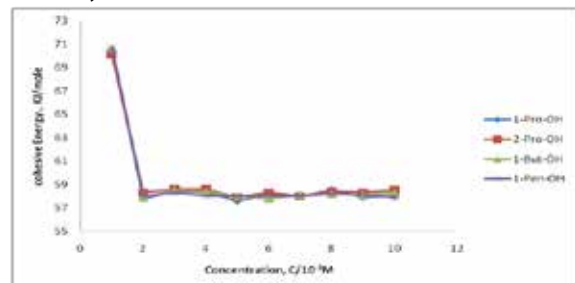**Figure 3. Plot of Internal Pressure Vs Concentration**
Solvent: Cyclohexane

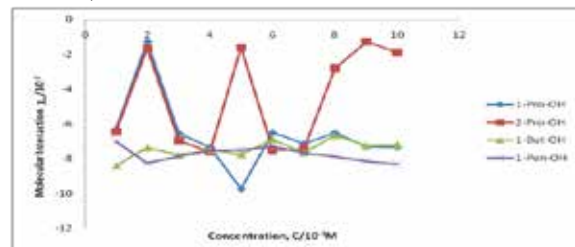**Figure 4. Plot of Free Length Vs Concentration**
Solvent: Cyclohexane

Figure 5. Plot of Cohesive Energy Vs Concentration
Solvent: Cyclohexane

Figure 6. Plot of Molecular interaction Vs Concentration
Solvent: Cyclohexane

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