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Ultrasonic Studies on the Molecular Interaction of Certain Aliphatic Dialdehyde Compounds with Ethylenediamine in n-Hexane at Different Temperature

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ABSTRACT

Acoustical parameters are calculated for three ternary systems of ethylenediamine and different dialdehydes such as glyoxal, glutaraldehyde, and succinaldehyde in n-hexane medium from the measurement of ultrasonic velocity, density, and viscosity at 303K, 308K, and 313K. From these data, acoustical parameters such as adiabatic compressibility, free length, free volume, internal pressure, absorption coefficient, viscous relaxation time, available volume, cohesive energy, Lenard-Jones potential, free energy of activation, formation constant, and molecular interaction parameter have been evaluated. From the measurements, investigated the complex formation through intermolecular hydrogen bonding between dialdehyde and ethylenediamine. Excess thermo acoustic parameters conclude the existence of hydrogen-bonded complexes between dialdehyde and amine.

Key words: *Dialdehyde, Ethylenediamine, Intermolecular hydrogen bond, Excess parameter.*

1. INTRODUCTION

Hydrogen bond gives the significance important for the formation complex and to analyze the strength of molecular interaction between solute and solvent. Amines have active hydrogen and are self-associated through intermolecular hydrogen bond. They have both π-electron as well as n-electron. Ultrasonic instrument has to be used in the detection of charge-transfer complexes [1,2] and in the calculation of stability constants and thermodynamic properties of the complexes [3-5]. Charge-transfer interaction plays an important in various fields [6-8]. Dialdehyde can form a strong complex with ethylenediamine, due to the presence of active hydrogen in these compounds. It is already reported that amines can form charge-transfer complexes with aldehydes and ketones [9,10]. The charge-transfer complexes of several aromatic aldehydes with amine have been studied by Kumar et al. [5]. The charge-transfer complexes of several aliphatic aldehydes with a different amine in n-hexane have been studied by Kannappan and Gandhi [11]. In the present work, three dialdehydes such as glyoxal, glutaraldehyde, and succinaldehyde were mixed separately with equimolar concentrations of ethylenediamine in n-hexane at different temperatures such as 303K, 308K, and 313K. The measurement of ultrasonic velocity, density, and viscosity has been made for this system. From these measurements, acoustical parameters were calculated. Thermodynamic parameters such as free energy formation and free energy of activation also evaluated using formation constant (K) values using a modified Bhatt equation proposed by Kannappan.

2. EXPERIMENTAL METHODS

The acceptors glyoxal, glutaraldehyde, succinaldehyde, and the donor ethylenediamine were AR samples. The non-polar solvent n-hexane was laboratory reagent which was purified by standard methods. The ultrasonic velocity of the pure liquid and their mixtures were calculated using ultrasonic interferometer (Mittal type:model:F81) working at a frequency of 2 MHZ with an overall accuracy of ±0.1 ms^{-1}. The density (ρ) can be measured using specific gravity bottles of capacity 10 ml. The viscosity (η) can be measured using Ostwald's viscometer with an accuracy of.0001 $Nm^{-2}s$. Acoustical parameters were calculated using standard equations [12,13-16]. Thermodynamical parameters can be calculated using modified Bhatt equation proposed by Kannappan [12,13].

$$K = Y/(b-y)^2$$

Where $Y = (a-K^{1/2})/(K-K^{1/2})$ in which K=x/y, K is the formation constant," 'b'is the difference between Ucal and Uobs at lower concentration 'a' " y is the difference between U_{cal} and U_{obs} at higher concentration b, and U_{cal} is the ultrasonic velocity of the mixture calculated

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from the mole fraction of the components using additive principle.

3. RESULTS AND DISCUSSION

3.1. Acoustical Parameters

Measured values of ultrasonic velocity (u), density (ρ), viscosity (η), and the calculated acoustical parameters for three ternary systems at 303K, 308K, and 313K were given in Tables 1-3. An inspection of the acoustical parameters in Tables 1-3 suggests that the measured properties of ultrasonic velocity (U) increase with increase in concentration for the three ternary systems. It can also proved by Figure 1. This property may be indicated to strong solute-solute interactions. It was also confirmed that the strength of molecular interactions in ternary liquid mixtures becomes high where the velocity maximum [17]. The polar nature of the dialdehyde may attribute the self-association of ethylenediamine. The gradual increase of ultrasonic velocity with concentration indicates the strong interaction such as complex formation or solute-solvent interaction through intermolecular hydrogen bond [18].

Adiabatic compressibility (β) shows the molecular association or dissociation. The intermolecular free length also supports this existence of association or dissociation. The decrease in adiabatic compressibility and intermolecular free length (L_f) with concentration proved the strength of interaction between the dialdehyde and ethylenediamine. It also supports for the compactness of the systems. The steep decrease of β leads the formation of strong coordinate systems. It also proved that the close packing within the molecules with decrease in free length [19].

Table 1: The value of ultrasonic velocity (u), density (ρ), viscosity (η), adiabatic compressibility (β), acoustical impedance (Z), free length (L_f), free volume (V_f), absorbtion coeffient, LJP, internal pressure (π_i), molar volume (V_a), and cohesive energy (CE) of n-hexane solution at 303K.

U	ρ	η	β	Z	L_f	V_f	AB.CO	LJP	π_i	V_a	CE
Glyoxal+ethylenediamine+n-hexane											
1031.6	939.8	0.888	1	9.69	6.22	3.58	2.26	3.89	1.625	3.26	14.9
1033.4	942.7	0.886	0.99	9.74	6.2	3.6	2.24	3.94	1.625	3.24	14.8
1035.2	945.8	0.884	0.98	9.79	6.18	3.63	2.22	4	1.625	3.22	14.8
1037.1	948.3	0.882	0.98	9.83	6.16	3.65	2.19	4.05	1.624	3.2	14.7
1039.7	951.2	0.88	0.97	9.89	6.13	3.68	2.16	4.13	1.623	3.18	14.7
1041.9	954.3	0.878	0.96	9.94	6.11	3.7	2.14	4.2	1.622	3.16	14.6
1043.8	957.5	0.876	0.95	9.99	6.09	3.73	2.12	4.26	1.622	3.13	14.6
1045.8	960.7	0.874	0.95	1	6.07	3.75	2.09	4.32	1.622	3.11	14.5
Succinaldehyde+ethylenediamine+n-hexane											
1045.3	876.4	0.748	1.04	9.16	6.36	4.72	1.96	4.31	1.414	3.41	13.9
1047.2	879.4	0.746	1.04	9.21	6.33	4.76	1.94	4.37	1.414	3.39	13.8
1049.3	882.5	0.744	1.03	9.26	6.31	4.79	1.92	4.43	1.414	3.36	13.8
1051.4	885.9	0.742	1.02	9.31	6.29	4.83	1.89	4.5	1.414	3.34	13.7
1053.9	888.2	0.74	1.01	9.36	6.26	4.87	1.87	4.58	1.413	3.32	13.7
1055.8	891.6	0.738	1.01	9.41	6.24	4.9	1.85	4.64	1.412	3.29	13.6
1057.7	894.3	0.736	1	9.46	6.22	4.94	1.83	4.7	1.411	3.27	13.6
1059.1	897.8	0.734	0.93	9.51	6.2	4.97	1.81	4.75	1.411	3.25	13.5
Glutaraldehyde+ethylenediamine+n-hexane											
1057.1	764.2	0.656	1.17	8.08	6.73	5.85	1.91	4.68	1.202	3.83	13.5
1059.7	767.2	0.654	1.16	8.13	6.7	5.9	1.88	4.77	1.202	3.83	13.5
1061.3	780.9	0.652	1.14	8.29	6.63	5.94	1.84	4.82	1.213	3.72	13.4
1063.5	783.1	0.65	1.13	8.33	6.61	5.99	1.81	4.89	1.211	3.69	13.3
1065.7	786.4	0.648	1.12	8.38	6.58	6.04	1.79	4.97	1.211	3.66	13.2
1067.3	789.2	0.646	1.11	8.42	6.56	6.08	1.77	5.02	1.211	3.64	13.2
1069.8	792.5	0.644	1.1	8.48	6.53	6.13	1.75	5.11	1.211	3.61	13.1
1071.4	795.9	0.642	1.09	8.53	6.51	6.18	1.72	5.16	1.211	3.58	13.1

LJP: Lenard-Jones potential

Table 2: The value of ultrasonic velocity (u), density (ρ), viscosity (η), adiabatic compressibility (β), acoustical impedance (Z), free length (L_f), free volume (V_f), absorbtion coeffient, LJP, internal pressure (π_i), molar volume (V_a), and cohesive energy (CE) of n-hexane solution at 308K.

U	P	η	β	Z	L_f	V_f	AB.CO	LJP	π_i	V_a	CE
Glyoxal+ethylenediamine+n-hexane											
1026.1	916.6	0.797	1.04	9.41	6.33	4.18	2.12	3.73	1.518	3.37	14.3
1028.9	919.4	0.795	1.03	9.46	6.31	4.21	2.09	3.83	1.517	3.35	14.2
1030.4	922.3	0.793	1.02	9.5	6.29	4.24	2.07	3.85	1.517	3.33	14.2
1032.4	925.5	0.791	1.01	9.55	6.26	4.27	2.04	3.91	1.516	3.31	14.1
1034.7	928.8	0.789	1.01	9.61	6.24	4.3	2.02	3.98	1.516	3.28	14.1
1036.6	931.3	0.787	0.99	9.65	6.22	4.33	1.99	4.04	1.515	3.26	14
1038.2	934.7	0.785	0.93	9.7	6.2	4.36	1.97	4.09	1.515	3.24	13.9
1040.5	937.1	0.783	0.86	9.75	6.18	4.39	1.95	4.16	1.513	3.22	13.9
Succinaldehyde+ethylenediamine+n-hexane											
1037.6	854.3	0.686	1.09	8.86	6.49	5.32	1.89	4.07	1.337	3.55	13.3
1039.4	857.1	0.684	1.08	8.91	6.46	5.36	1.87	4.12	1.336	3.52	13.3
1041.3	860.9	0.682	1.07	8.98	6.44	5.4	1.84	4.18	1.336	3.5	13.2
1043.6	863.5	0.68	1.06	9.01	6.41	5.44	1.82	4.25	1.336	3.47	13.2
1045.8	866.7	0.678	1.05	9.06	6.39	5.48	1.8	4.32	1.335	3.45	13.1
1047.2	869.2	0.676	1.05	9.1	6.37	5.52	1.78	4.37	1.334	3.43	13.1
1049.2	871.8	0.674	1.04	9.15	6.35	5.56	1.76	4.43	1.333	3.41	13
1051.7	874.6	0.672	1.03	9.2	6.32	5.61	1.74	4.51	1.332	3.38	13
Glutaraldehyde+ethylenediamine+n-hexane											
1049.3	717.9	0.613	1.27	7.53	7	6.4	1.94	4.43	1.119	4.13	13.4
1051.4	720.8	0.611	1.26	7.58	6.97	6.45	1.92	4.5	1.118	4.1	13.4
1053.7	723.5	0.609	1.24	7.62	6.94	6.51	1.89	4.57	1.118	4.07	13.3
1055.7	726.6	0.607	1.23	7.67	6.91	6.56	1.87	4.64	1.118	4.04	0.3
1057.9	729.1	0.605	1.23	7.71	6.89	6.62	1.84	4.71	1.117	4.01	13.2
1059.4	732.4	0.603	1.22	7.76	6.86	6.67	1.82	4.76	1.117	3.98	13.2
1061.4	735.7	0.601	1.21	7.81	6.83	6.72	1.8	4.82	1.117	3.95	13.1
1063.6	738.9	0.599	1.2	7.86	6.8	6.78	1.77	4.9	1.117	3.92	13.1

LJP: Lenard-Jones potential

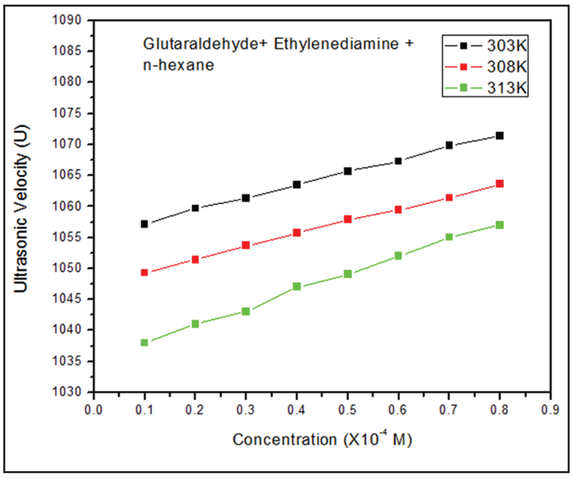

Figure 1: Plots of ultrasonic velocity versus various concentration of glutaraldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

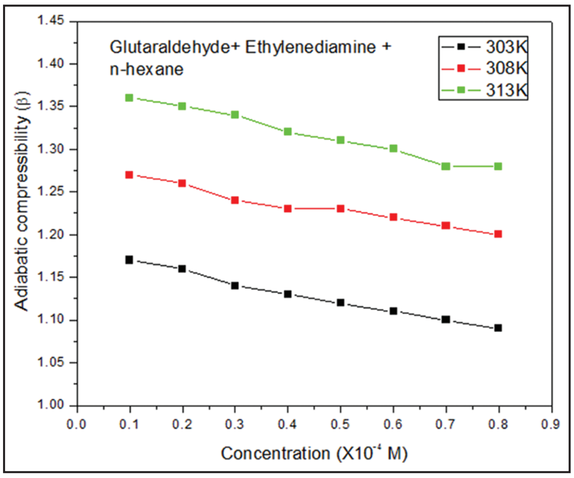

Figure 2: Plots of adiabatic compressibility versus various concentrations of glutaraldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

Table 3: The value of ultrasonic velocity (u), density (ρ), viscosity (η), adiabatic compressibility (β), acoustical impedance (Z), free length (L_f), Free volume (V_f), absorbtion coeffient, LJP, internal pressure (π_i), molar volume (V_a), and cohesive energy (CE) of n-hexane solution at 313K.

U	ρ	η	β	Z	L_f	V_f	AB.CO	LJP	π_i	V_a	CE
Glyoxal+ethylenediamine+n-hexane											
1019.4	904.5	0.723	1.06	9.22	6.42	4.79	1.98	3.53	1.438	3.46	13.6
1021.4	907.8	0.721	1.06	9.27	6.39	4.82	1.96	3.59	1.438	3.43	13.6
1023.6	910.4	0.719	1.05	9.32	6.37	4.86	1.94	3.66	1.436	3.41	13.6
1025.9	913.4	0.717	1.04	9.37	6.34	4.9	1.91	3.72	1.436	3.39	13.5
1027.4	916.9	0.715	1.03	9.42	6.32	4.93	1.89	3.77	1.436	3.37	13.5
1029.2	919.2	0.713	1.03	9.46	6.3	4.97	1.87	3.82	1.434	3.35	13.4
1031.3	921.6	0.711	1.02	9.5	6.28	5.01	1.85	3.88	1.433	3.33	13.4
1033.5	924.7	0.709	1.01	9.56	6.26	5.04	1.83	3.95	1.432	3.31	13.4
Succinaldehyde+ethylenediamine+n-hexane											
1028.1	821.6	0.656	1.15	8.45	6.68	5.61	1.93	3.79	1.27	3.75	13.4
1030.9	824.4	0.654	1.14	8.5	6.65	5.66	1.9	3.87	1.27	3.72	13.4
1032.6	827.4	0.652	1.13	8.54	6.62	5.7	1.88	3.92	1.27	3.7	13.3
1034.7	839.4	0.65	1.11	8.69	6.56	5.75	1.84	3.98	1.27	3.63	13.1
1036.8	842.4	0.648	1.1	8.73	6.54	5.79	1.81	4.05	1.26	3.61	13.07
1038.4	845.6	0.646	1.1	8.78	6.51	5.84	1.79	4.09	1.26	3.58	13.02
1040.2	848.2	0.642	1.09	8.82	6.49	5.91	1.77	4.15	1.23	3.56	12.9
1042.6	851.2	0.64	1.08	8.87	6.47	5.96	1.74	4.22	1.22	3.53	12.9
Glutaraldehyde+ethylenediamine+n-hexane											
1038	681.3	0.599	1.36	7.07	7.26	6.52	2.07	4.08	1.075	4.44	13.5
1041	684.6	0.597	1.35	7.13	7.22	6.58	2.03	4.17	1.074	4.4	13.5
1043	687.4	0.595	1.34	7.17	7.19	6.64	2.01	4.24	1.073	4.37	13.4
1047	690.2	0.593	1.32	7.23	7.15	6.71	1.97	4.36	1.072	4.32	13.4
1049	693.7	0.591	1.31	7.28	7.12	6.77	1.94	4.42	1.072	4.28	13.3
1052	696.3	0.589	1.3	7.33	7.09	6.84	1.91	4.52	1.071	4.25	13.3
1055	699.7	0.587	1.28	7.38	7.05	6.9	1.88	4.61	1.071	4.2	13.2
1057	701.5	0.585	1.28	7.41	7.03	6.96	1.86	4.68	1.07	4.18	13.1

LJP: Lenard-Jones potential

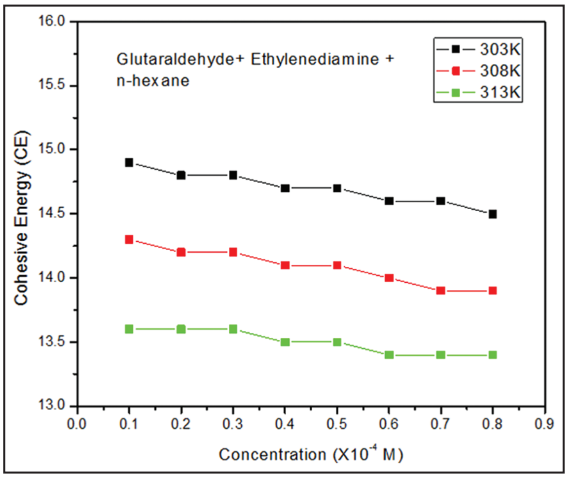

Figure 3: Plots of cohesive energy versus various concentrations of glutaraldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

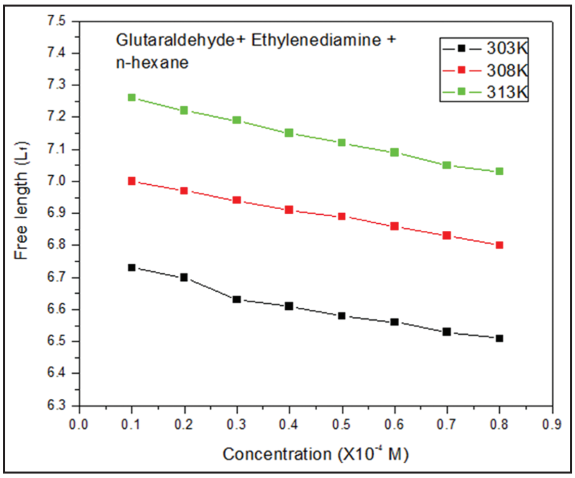

Figure 4: Plots of free length versus various concentrations of glutaraldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

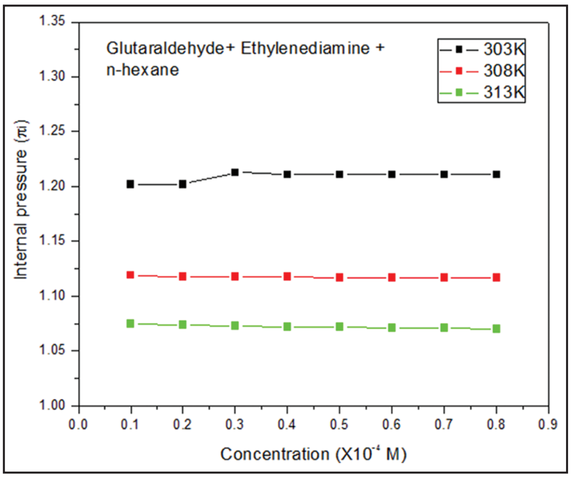

Figure 5: Plots of internal pressure versus various concentrations of glutaraldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

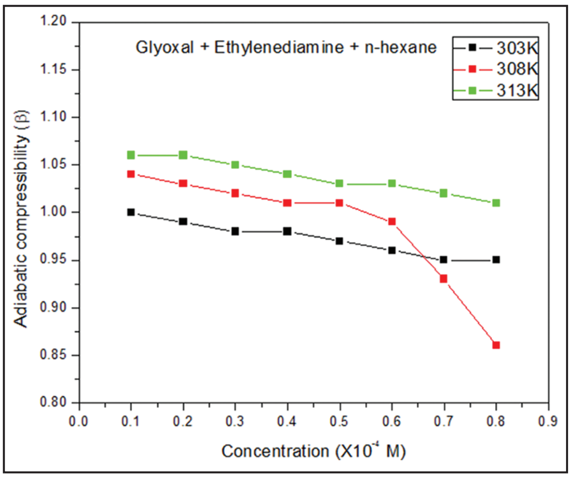

Figure 6: Plots of adiabatic compressibility versus various concentrations of glyoxal - ethylenediamine in n-hexane at 303K, 308K, and 313K.

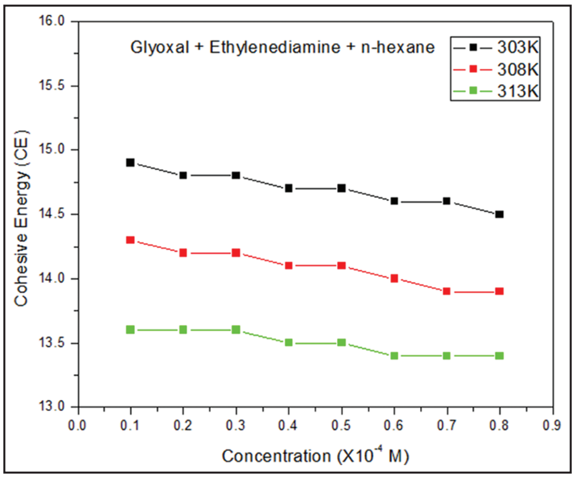

Figure 7: Plots of cohesive energy versus various concentrations of glyoxal - ethylenediamine in n-hexane at 303K, 308K, and 313K.

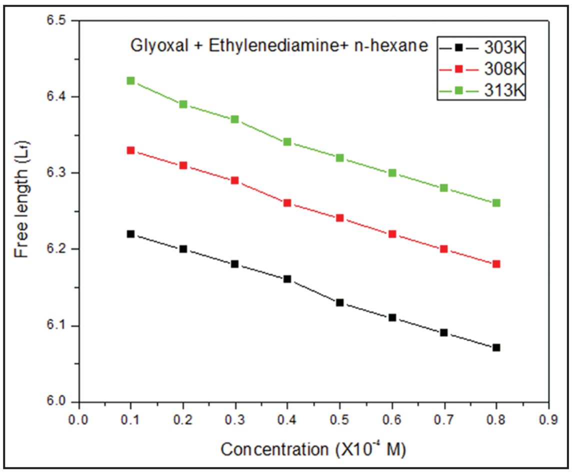

Figure 8: Plots of free length versus various concentrations of glyoxal - ethylenediamine in n-hexane at 303K, 308K, and 313K.

The force of attraction and force of repulsion can be proved by the measure of internal pressure (π_i). The internal pressure increases with the concentration of all the systems. From Tables 1-3, it showed the force of attraction and strong dipole-dipole interaction from the internal pressure values. The reverse trend of free volume revealed the strong coordination between dialdehyde and ethylenediamine in n-hexane medium. It also suggests the strong solute-solute interaction through intermolecular hydrogen bond [17] (Figure 2-15).

The cohesive energy shows intermolecular hydrogen and the dipole-dipole interaction. In the present study, there is no significant change of cohesive energy. The small and gradual variation was observed. The magnitude of cohesive energy is gradual changes or almost constant for all the system. The variation of Lenard-Jones potential (LJP) is almost similar for all the three systems. The small variation and gradual increasing of LJP show the presence of hydrogen bonding and complex formation in the ternary liquid mixtures.

3.2. Analysis of Thermodynamic Parameter

Among saturated aliphatic dialdehydes, the stability increases with increases in the length of alkyl group. So that glutaraldehyde forms more stable complex with amine than the succinaldehyde and glyoxal. The formation constant (K) can be used to determine the stability of charge-transfer complexes [12,13]. This parameter is also used to assess the strength of molecular attraction between the donor and acceptor. The formation constant is concentration dependent and is presented in Table 4 because these values increase with concentration. The K value indicates that the stability of complexes depends on the structure of donor-acceptor. In general, aliphatic aldehyde forms more stable complex than aromatic aldehydes.

Because the positive charge decreases on phenyl ring due to mesmeric and inductive effects [11]. The free energy of activation ($\Delta G^{\#}$) and relaxation time values were presented in Tables 5 and 6. The same value of free energy activation and relaxation time indicates that similar type of complexes formed between the dialdehyde and amine. The negative values of molecular interaction parameter also show the presence of strong interaction between carbonyl compounds and ethylenediamine. It can be represented in Table 7.

3.3. Analysis of Excess Parameter

The existence of any intermolecular interaction can be explained using the excess values of the thermodynamical parameters. The strength of interaction depends on the sign of excess values [20,21].

Table 4: Formation constant K for glyoxal, succinaldehyde, and glutaraldehyde with ethylenediamine in n-hexane at 303K, 308K, and 313K.

Concentration×10^{-4}	**Glyoxal**			**Succinaldehyde**			**Glutardehyde**		
	303K	**308K**	**313K**	**303K**	**308K**	**313K**	**303K**	**308K**	**313K**
1	164.3	225.5	148.5	230.1	184.4	232.9	421.6	278.8	296.5
2	161.4	126.6	159.2	244	189.6	146.8	277	290.8	204.7
3	165.9	159.9	162.4	237.2	216.1	172.6	333.4	253.8	331.3
4	206.2	175.1	112.2	258.6	204	169.2	318.4	261.5	203.1
5	173.6	143	122.6	204.5	136	128.5	250.3	190.8	248.8
6	158.8	129	141.3	205.3	179.7	144.6	296.7	227.9	243.8
7	160.7	161.4	143.2	168.4	198.1	170.2	244.8	229.4	194.4
8									

Table 5: Free energy of activation $\Delta G^{\#}$ KJ/mole for glyoxal, succinaldehyde, and glutaraldehyde with ethylenediamine in n-hexane at 303K, 308K, and 313K.

Concentration×10^{-4}	**Glyoxal**				**Succinaldehyde**			**Glutaraldehyde**	
	303K	**308K**	**313K**	**303K**	**308K**	**313K**	**303K**	**308K**	**313K**
1	2.46	2.44	2.42	2.43	2.42	2.42	2.42	2.42	2.44
2	2.46	2.44	2.42	2.42	2.41	2.42	2.42	2.42	2.44
3	2.46	2.44	2.42	2.42	2.41	2.41	2.41	2.42	2.43
4	2.46	2.44	2.42	2.42	2.41	2.41	2.41	2.42	2.43
5	2.45	2.43	2.41	2.42	2.4	2.4	2.41	2.41	2.42
6	2.45	2.43	2.41	2.41	2.4	2.4	2.4	2.41	2.42
7	2.45	2.43	2.41	2.41	2.4	2.4	2.4	2.41	2.42
8	2.45	2.42	2.4	2.41	2.4	2.4	2.4	2.4	2.42

Table 6: Viscosous relaxation time r for glyoxal, succinaldehyde, and glutaraldehyde with ethylenediamine in n-hexane at 303K, 308K, and 313K.

Concentration×10^{-4}	**Glyoxal**			**Succinaldehyde**			**Glutaraldehyde**		
	303K	**308K**	**313K**	**303K**	**308K**	**313K**	**303K**	**308K**	**313K**
1	1.18	1.1	1.03	1.04	0.99	1.01	1.02	1.03	1.09
2	1.17	1.09	1.02	1.03	0.98	0.95	1.01	1.02	1.07
3	1.16	1.08	1.01	1.02	0.97	0.98	0.98	1.01	1.06
4	1.15	1.07	0.99	1.01	0.96	0.96	0.97	0.99	1.05
5	1.14	1.06	0.98	1	0.95	0.95	0.96	0.98	1.03
6	1.13	1.05	0.97	0.99	0.94	0.94	0.95	0.97	1.02
7	1.12	1.04	0.96	0.98	0.93	0.93	0.94	0.96	1
8	1.11	1.03	0.95	0.97	0.92	0.92	0.93	0.95	0.99

The excess values present in Tables 8-10. The excess ultrasonic velocity is negative at low concentration, and it is positive at high concentration which can be concluded the high interaction at high concentration. The negative values of excess adiabatic compressibility and excess free length show the strong interaction in liquid mixtures. The negative sign of excess free length becomes a compactness due to molecular interaction through dipole-dipole interaction [22,23], indicating high compact structure making which enhances excess free length to have negative values. The positive value of excess impedance and excess velocity at high concentration suggests the presence of strong interaction in the components mixture [24,25]. The close packing conforms that the trend in excess free volume and excess internal pressure is opposite to each other. Excess free volume is negative, and excess internal pressure is positive.

Table 7: Molecular interaction parameter χu/10-2 for glyoxal, succinaldehyde, and glutaraldehyde with ethylenediamine in n-hexane at 303K, 308K, and 313K.

Concentration×10^{-4}	Glyoxal			Succinaldehyde			Glutaraldehyde		
	303K	308K	313K	303K	308K	313K	303K	308K	313K
1	−0.002	−0.0026	−0.0028	0.00182	−0.00096	−0.0009	−0.0035	−0.002	−0.0017
2	−0.0052	−0.0097	−0.0087	−0.0064	−0.005	−0.0009	−0.0109	−0.0058	−0.0081
3	−0.0082	−0.0122	−0.0125	−0.0095	−0.0078	−0.0025	−0.0131	−0.0099	−0.0111
4	−0.0112	−0.0147	−0.0157	−0.0127	−0.0113	−0.0059	−0.0163	−0.0127	−0.0173
5	−0.0149	−0.0187	−0.0176	−0.0157	−0.0149	−0.0094	−0.0185	−0.0138	−0.0199
6	−0.018	−0.0196	−0.0199	−0.0179	−0.0148	−0.0123	−0.0205	−0.0147	−0.024
7	−0.0202	−0.0205	−0.0233	−0.0186	−0.0163	−0.0142	−0.0225	−0.0169	−0.0282
8	−0.0213	−0.0233	−0.026	−0.0201	−0.0189	−0.0168	−0.0241	−0.0193	−0.0289

Table 8: Excess values ultrasonic velocity (U^E), excess adiabatic compressibility (β^E), excess free length ($L^E{}_f$), excess acoustical impedance (Z^E), excess free volume (V_f^E), and excess internal pressure ($\pi^E{}_f$) for glyoxal with ethylenediamine in n-hexane medium at 303K.

Concentration×10^{-4}	U^E	β^E	$L^E{}_f$	ZE	V_f^E	π_f^E
1	−71.57	−0.905	−48.2	867088.1	2	2.97
2	−39.37	−0.934	−12.8	861037.5	−6.4	3.71
3	4.6	−0.95	−4.01	864997	−6.8	3.71
4	19.42	−0.966	−2.32	860963.8	−8.4	3.73
5	54.2	−0.982	−1.11	862936.2	−4.3	3.69
6	73.85	−0.998	−0.575	859912.8	−1.5	3.76
7	91.13	−1.01	−0.338	864892.7	−0.66	3.72
8	104.73	−1.03	−0.703	861875.4	−0.2	3.69

Table 9: Excess values ultrasonic velocity (U^E), excess adiabatic compressibility (β^E), excess free length ($L^E{}_f$), excess acoustical impedance (Z^E), excess free volume (V_f^E), and excess internal pressure ($\pi^E{}_f$) for succinaldehyde with ethylenediamine in n-hexane medium at 303K.

Concentration×10^{-4}	U^E	β^E	$L^E{}_f$	ZE	V_f^E	π_f^E
1	−84.67	−0.909	−85.7	867126.3	2	2.97
2	−59.18	−0.925	−22.7	861103.1	−6.4	3.71
3	−17.89	−0.937	−7.13	865083	−6.8	3.71
4	−3.36	−0.95	−4.12	861065.2	−8.4	3.73
5	32.67	−0.962	−1.97	863049.5	−4.3	3.69
6	54.53	−0.974	−0.601	860035.5	−1.5	3.77
7	74.61	−0.987	−0.33	865022.9	−0.66	3.73
8	91.35	−0.999	−0.125	862011.5	−0.2	3.69

Table 10: Excess values ultrasonic velocity (U^E), excess adiabatic compressibility (β^E), excess free length ($L^E{}_f$), excess acoustical impedance (Z^E), excess free volume (V_f^E), and excess internal pressure ($\pi^E{}_f$) for glutaraldehyde with ethylenediamine in n-hexane medium at 303K.

Concentration×10^{-4}	U^E	β^E	$L^E{}_f$	ZE	V_f^E	π_f^E
1	−87.53	−0.92	−99.7	867128	2	2.97
2	−63.81	−0.932	−26.4	861106	−6.4	3.71
3	−23.54	−0.945	−8.29	865086.5	−6.8	3.71
4	−9.48	−0.957	−4.79	861069	−8.4	3.73
5	26.46	−0.969	−2.29	863053.3	−4.3	3.69
6	48.51	−0.982	−1.19	860039.2	−1.5	3.77
7	68.99	−0.994	−0.699	865026.3	−0.66	3.73
8	86.27	−1	−0.384	862014.6	−0.2	3.7

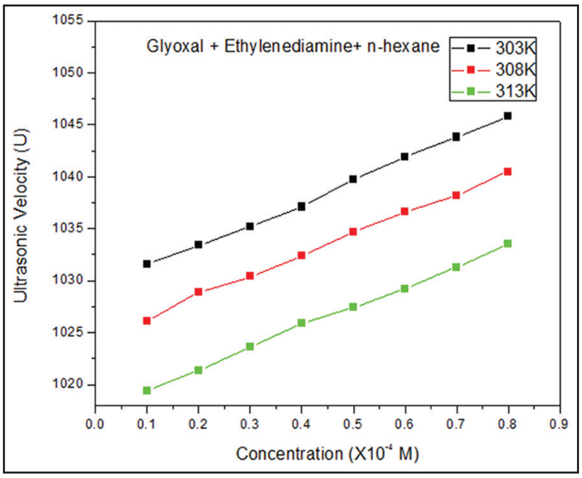

Figure 9: Plots of ultrasonic velocity versus various concentrations of glyoxal - ethylenediamine in n-hexane at 303K, 308K, and 313K.

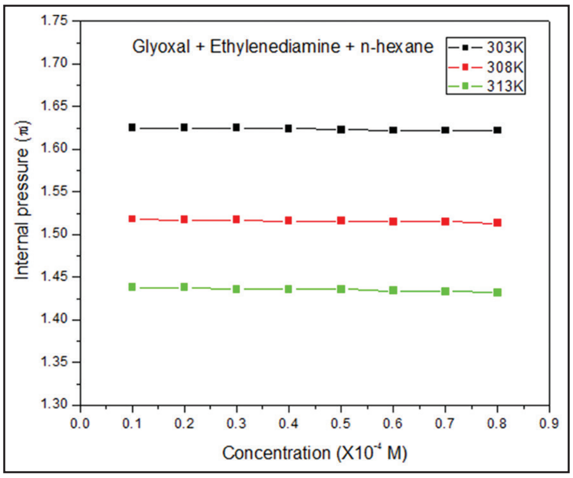

Figure 10: Plots of internal pressure versus various concentration of glyoxal - ethylenediamine in n-hexane at 303K, 308K, and 313K.

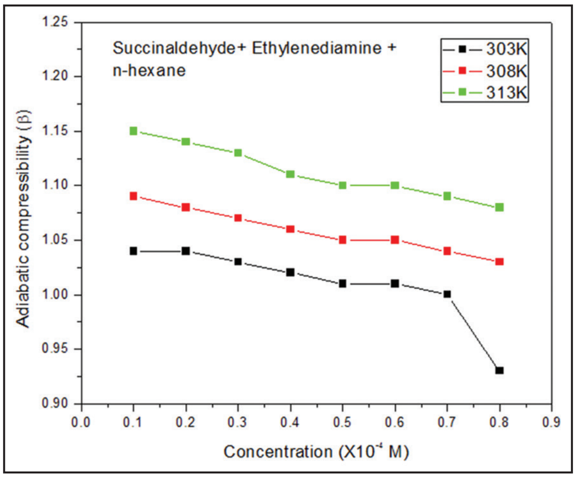

Figure 11: Plots of adiabatic compressibility versus various concentrations of succinaldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

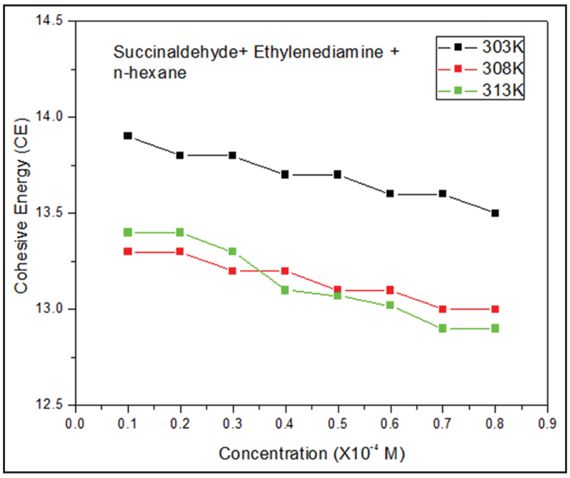

Figure 12: Plots of cohesive energy versus various concentration of succinaldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

4. CONCLUSION

The solute-solvent interaction through intermolecular hydrogen bond is strong with the evidence of increasing ultrasonic velocity values in ternary system. The charge-transfer complex formed between

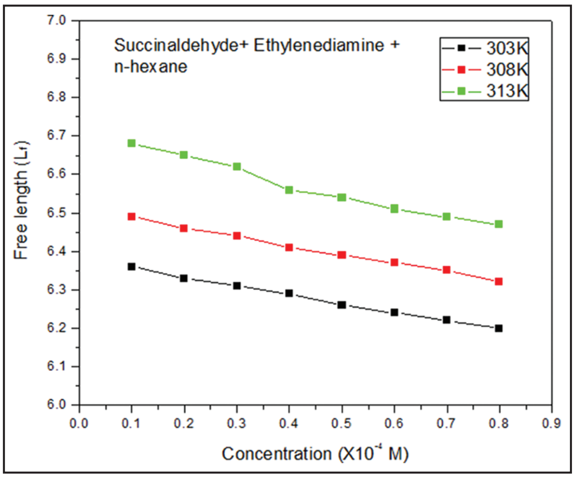

Figure 13: Plots of free length versus various concentrations of succinaldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

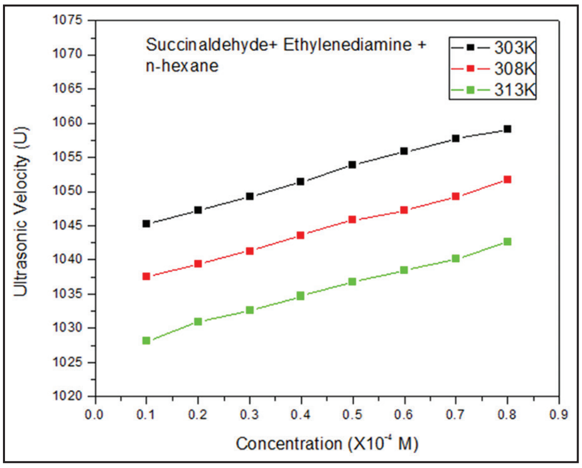

Figure 14: Plots of ultrasonic velocity versus various concentration of succinaldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

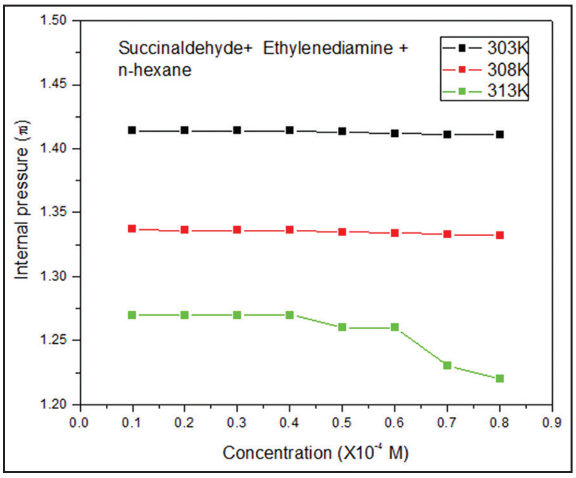

Figure 15: Plots of internal pressure versus various concentration of succinaldehyde - ethylenediamine in n-hexane at 303K, 308K, and 313K.

three dialdehydes with ethylenediamine in n-hexane. It can be proved by the determination of acoustical parameters and thermodynamic parameters. The stability is in the order glyoxal-ethylenediamine < succinaldehyde-ethylenediamine < glutaraldehyde-ethylenediamine. This order is proved by K values. The complexes are thermodynamically stable as proved from their negative free energy of formation values. The complexes are structurally similar type as evidenced their constant value of relaxation time. The excess values also support the presence of molecular interaction in the liquid mixture.

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