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Thermodynamic and volumetric properties of hydroxamic acids in dimethylsulfoxide at $T = (298.15$ to $313.15)$ K

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ABSTRACT

Physical properties, such as density, (ρ) and refractive index, (n) of three hydroxamic acids (*N*-phenylbenzo-, *N*-phenyl-4-methyl-3-nitrobenzo-, and *N*-phenyl-4-nitrobenzo-) were measured in dimethyl sulfoxide (DMSO) as a function of concentrations at $T = (298.15, 303.15, 308.15,$ and $313.15)$ K . The apparent molar volumes (V_ϕ^0), limiting apparent molar volumes (V_ϕ^0) at infinite dilution, slope (S_V^*) and Hepler's constant ($\partial^2 V_\phi^0 \partial T^2$) are calculated from the experimental values of densities (ρ) by applying the Masson's equation. The apparent molar expansibilities at infinite dilution ($\phi^0{}_E$), molar volumes (V), thermal expansion coefficient (α_2) and the excess molar volumes (V^E) are also computed. The precise refractive indices (n) data have been used to evaluate the steric parameters viz. molar refractions (R_M), polarizability (α) and excess molar refraction (R_M^E) of these molecules. It is inferred from these results that the above mentioned drugs act as structure-making compound due to hydrophobic hydration of the molecules in the drug.

1. Introduction

Volumetric and optical properties are good candidates for the study of molecular interaction and extension of solution theory because they depend on solute–solute and solute–solvent interactions and the structural effects arising from interstitial accommodation [1–5]. Properties such as densities, refractive indices and their variation with temperature and composition of the solution are useful to design engineering processes and in chemical and biological industries [6–10]. The parameters, apparent molar volumes and limiting apparent molar volumes of dilute solutions can be used for the development of molecular models for describing the thermodynamic behavior of solutions. The V_ϕ^0 depends upon molecular size, shape, interactions and structural effects among the solvent [11]. Excess properties of solutions, such as deviation in molar refraction called excess molar refraction, R_M^E and V^E are useful for the design of separation techniques and to test theories of solutions [12].

Dimethylsulfoxide (DMSO) a tipical aprotic solvent having both polar and nonpolar groups, isan important solvent in chemistry, biotechnology, and medicine for the dissolution of various substance and as an antidepressing agent of living cell. The hydroxamic acid functionality, –C(=O).N.OH, is a key structural constituent of many biomolecules, some of which, are naturally occurring and others, such as peroxidase, matrix metalloproteinase and urease inhibitors [13,14] are of synthetic origin. Hydroxamic acid derivatives have received increasing attention due to their biological activity especially as enzyme inhibitors [15] and metal chelators [16]. Hydroxamic acids are versatile reagents in analytical chemistry [17,18] and are widely used in medicine as analgetics, anti-inflamatories [19], antibiotics [20], anticancer agent [21], antifungal and hypotentive agent [22].

Over the past decade, our research group has made some remarkable efforts to study the steric and excess properties of binary and pure systems of different derivatives of hydroxamic acids. The data are lacking in the literature on the densities and optical properties of PBHA, PMNHA, and PNHA in pure DMSO at different temperatures. Therefore, in the present paper, we report ρ, n of solution over entire range of concentration and at temperatures, $T = (298.15, 303.15, 308.15,$ and $313.15)$ K. These data are further used to calculate V_ϕ^* , V_ϕ^0, $\phi^0{}_E$, V^E, n^E and R_M, in order to understand molecular behavior and the nature of solute-solvent interactions [23–25].

2. Experimental

2.1. Materials

Three hydroxamic acids namely *N*-phenylbenzohydroxamic acid (PBHA), *N*-phenyl-4-methyl-3-nitrobenzohydroxamic acid (PMNHA) and *N*-phenyl-4-nitrobenzohydroxamic acid (PNHA) were prepared by the procedure reported in literature [26]. The hydroxamic acids were then purified by crystallizing thrice with benzene and dried over phos-

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phorus pentoxide in vacuum for several hours. The purity of the compounds was ascertained by determining their melting points, UV, IR spectra, LC-MS and element analysis. Melting point were determined with melting point apparatus (TEMPO) and are uncorrected. The uncertainty of measurement was ±1°C. Elemental analysis were performed with a Vario-EL analysis apparatus. IR spectra were recorded with a FTIR 8400 series Shimazdu (Japan) using KBr pellets. The data were tallied with the literature [27]. The LC–MS validated method can be applied as a generic method to detect the hydroxamic acid for pharmaceutical process control and drug substance release. DMSO, Merck (mass fraction purity>0.99) was used without further purification. Stock solution of PBHA (0.2345 M), PMNHA (0.1838 M), and PNHA (0.1936 M) were prepared in DMSO. Solutions of varying concentrations were then obtained from the stock solution by mass dilution technique. Uncertainties in solution concentration were estimated to be ±0.001 unit. The characteristics of synthesized compounds and specification of the purity of sample are given in Table 1.

2.2. Measurement of density, ρ

Densities of DMSO and hydroxamic acid solutions were determined using a 10^3 cm^3 double-armed pycnometer at temperatures T = (298.15, 303.15, 308.15, and 313.15) K. The pycnometer was calibrated thrice at each temperature with freshly prepared triple distilled water and data were averaged. All the weighing were performed on single pan balance (Sartorius BS 223S) with a precision of ±0.01mg. A thermostatically controlled well stirred bath (MaHaRaNa, Model No. IMC 732B, instruments Mfg. company, Ajmer, India), with a temperature controlled to ± 0.01K was used. The estimated precision of the density measurement of solution is ± 3×10^{-4} kg.m^{-3} and the uncertainty in the density measurement is ± 2×10^{-4} kg.m^{-3}.

2.3. Measurement of refractive index, n

Steric parameters were measured as refractive indices with a thermostated Abbe's refractometer as a function of solutes' concentration at T = (298.15, 303.15, 308.15, and 313.15) K. The refractometer was calibrated by measuring the refractive indices of triple distilled water and solvent at known temperature. The temperature of the thermostated water bath (JULABO. Germony) was maintained within ±0.02 K. The sample solutions were directly injected into prism assembly by means of an airtight hypodermic syringe. The error in refractive index measurement is less than ± 0.0001 units. Table 2 reports the densities and refractive indices of DMSO and hydroxamic acids.

3. Results and discussion

The ρ and n values of PBHA, PMNHA, and PNHA in DMSO at T = (298.15, 303.15, 308.15, and 313.15) K are presented as a function of their concentrations in Table 3 . Figs. 1 and 2 show that ρ and n increases with increase in the concentration of hydroxamic acids respectively.

3.1. Volumetric studies

The experimental values of solutions' ρ are used to calculate the V_ϕ^* by means of the following expression [38]:

$$V_\phi = M_2/\rho_0 + \left[10^3(\rho_0 - \rho)/Cr_0\right] \quad (1)$$

where, V_ϕ is the apparent molar volumes, C is the molarity of solute in the solutions, M_2 is the molar mass of the solute and ρ_0 and ρ are the densities of DMSO and the solutions, respectively. The values of V and V_ϕ^* are listed in Table 3. The positive values of V_ϕ for all the molecules indicate strong solute-solvent interactions. These interactions are strengthened with increasing concentration at a particular temperature and are

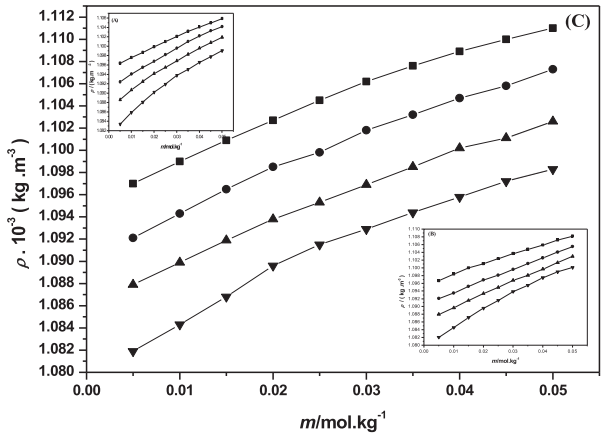


Fig. 1. Plot of densities (ρ) vs concentration (M) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures: ■, 298.15 K; ●, 303.15 K; ◄, 308.15 K; and ►, 313.15 K.

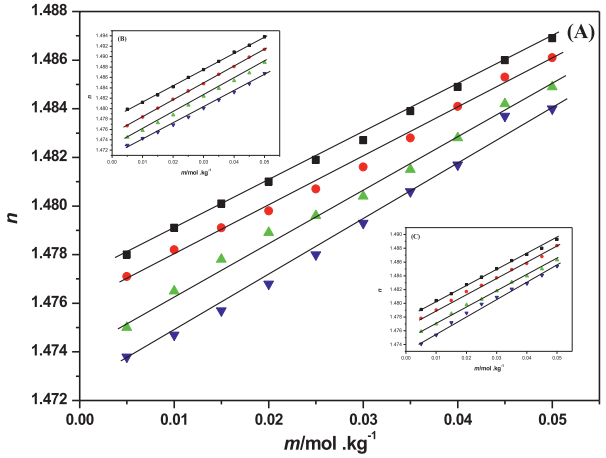


Fig. 2. Plot of refractive indices (n) vs concentration (M) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures: ■, 298.15 K; ●, 303.15 K; ◄, 308.15 K; and ►, 313.15 K.

weakened with a rise in temperature at a constant solutes' concentration as shown in Fig. 3. The apparent molar volume at infinite dilution is calculated by a least-squares treatment of the plot of V_ϕ versus $C^{1/2}$ using Masson's expression [38]:

$$V_\phi = V_\phi^0 + S_V^* C^{1/2} \quad (2)$$

where, V_ϕ^0 is the apparent molar volume at infinite dilution and S_V^* is the experimental slope. The data are listed in Table 4. V_ϕ^0 is regarded as resulting from solute-solvent interactions. The positive values indicate the presence of strong solute-solvent interactions. These interactions are weakened with a rise in temperature as shown in Fig. 4.

The temperature dependence of V_ϕ^0 [39] for the studied compounds is expressed by the equation:

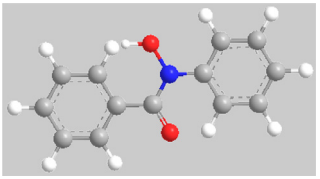
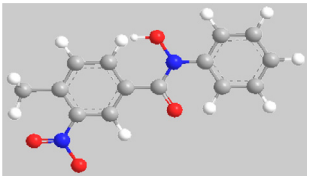
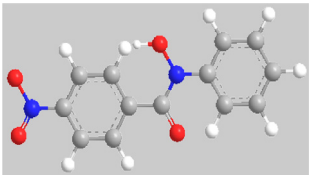
$$V_\phi^0 = a + bT + cT^2 \quad (3)$$

The temperature T is expressed in Kelvin. The coefficients a, b, and c have been estimated by the least-square fitting of the V_ϕ^0 in Eq. (3) and the following equations are formulated:

$$V_\phi^0 = 865.5083(\pm 7862.377) - 1.842(\pm 51.460)T - 0.002(\pm 0.084)T^2 \text{for(PBHA)} \quad (4)$$

$$V_\phi^0 = -8998.125(\pm 5954.738) + 63.002(\pm 38.974)T + 0.108(\pm 0.063)T^2 \text{for(PMNHA)} \quad (5)$$

Table 1
Characteristics of hydroxamic acids.

S. No.	Hydroxamic Acids	3D Structure	Melting temperature		IR (cm^{-1})				Elemental analyses					
					Observed				Observed			Theoretical		
			Observed	Reported	υ (O-H) cm^{-1}	υ (C=O) cm^{-1}	υ (C-N) cm^{-1}	υ (N-O) cm^{-1}	C	H	N	C	H	N
I	***N*– phenylbenzohydroxamic acid, (PBHA) ($C_{13}H_{11}NO_2$) *M*=213.23**		122°C	121°C[a]	3100	1642	1340	915	73.29	4.91	6.46	73.23	5.20	6.57
II	***N*–phenyl-4-methyl nitrobenzohydroxamic acid, (PMNHA) ($C_{14}H_{12}N_2O_4$) *M* =272.26**		117°C	117°C[b] 119°C[c]	3080	1647	1350	946	61.67	4.04	9.93	61.76	4.44	10.29
III	***N*-phenyl-4-nitrobenzohydroxamic acid, (PNHA) ($C_{13}H_{10}N_2O_4$) *M* =258.23**		168°C	168°C[a]	3186	1612	1349	916	60.46	3.81	10.73	60.47	3.90	10.85

[a] Ref. [28].
[b] Ref. [29].
[c] Ref. [30].

Table 2
Properties of DMSO and hydroxamic acids at different temperatures.

T/K	DMSO				Hydroxamic Acids					
	ρ_0. 10^{-3} (kg.m^{-3})		n_o		ρ_1. 10^{-3} (kg.m^{-3})			n_1		
	(Exptl.)	(Lit.)	(Exptl.)	(Lit.)	I	II	III	I	II	III
298.15	1.0949	1.0955[a]	1.4770	1.4771[e]	1.0956	1.0959	1.0953	1.4768	1.4773	1.4775
		1.0948[b]		1.4765[f]						
		1.0954[c]		1.4775[e]						
303.15	1.0898	1.0896[d]	1.4753	1.4752[e]	1.0906	1.0910	1.0902	1.4756	1.4758	1.4762
		1.0908[b]		1.4740[f]						
308.15	1.0855	1.0855[a]	1.4730	1.4720[f]	1.0862	1.0869	1.0861	1.4743	1.4745	1.4740
		1.0847[d]								
		1.0861[b]								
313.15	1.0793	1.0797[d]	1.4710	1.4492[e]	1.0801	1.0809	1.0799	1.4725	1.4730	1.4730
		1.0805[b]		1.4700[g]						

[a] Ref. [31].
[b] Ref. [32].
[c] Ref. [33].
[d] Ref. [34].
[e] Ref. [35].
[f] Ref. [36].
[g] Ref. [37].

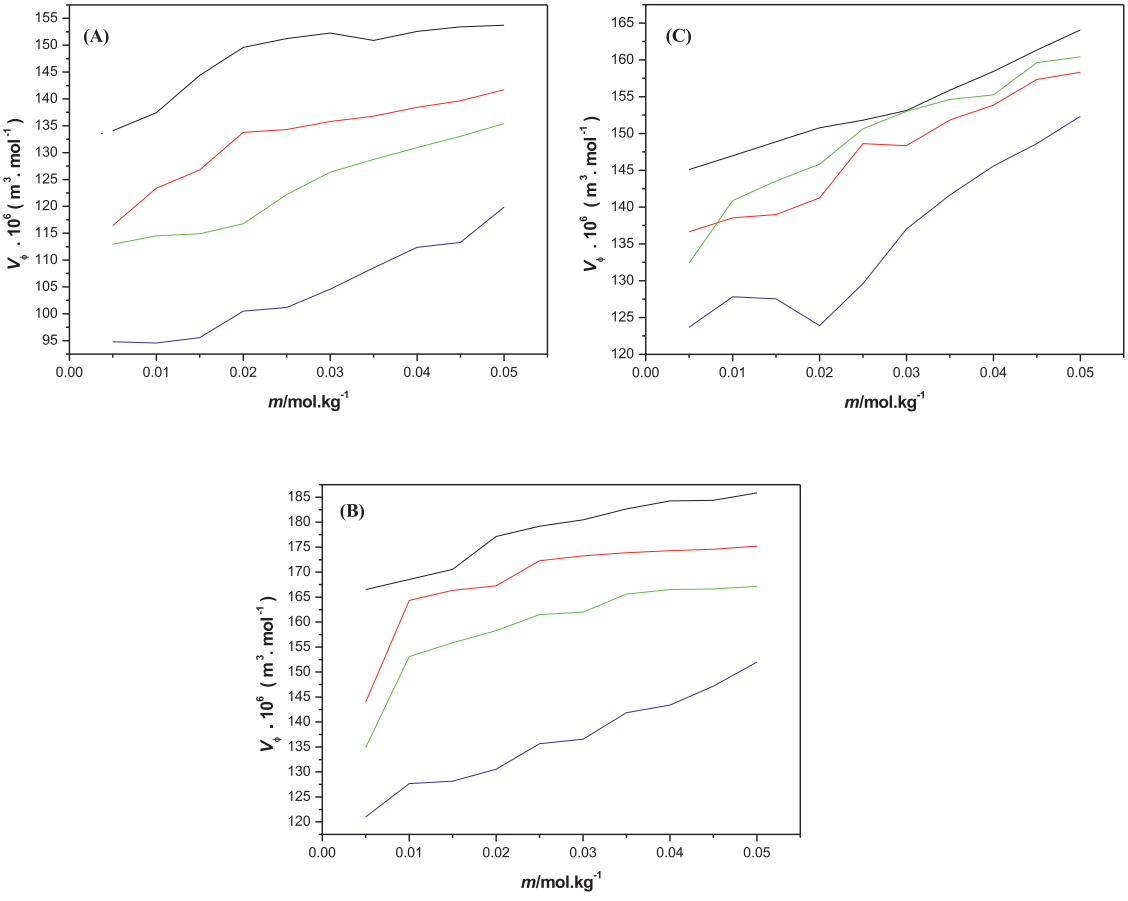


Fig. 3. Plot of apparent molar volume (V_ϕ) vs concentration (M) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures:, 298.15 K;, 303.15 K;, 308.15 K; and, 313.15 K.

$$V_\phi^0 = -3800.856(\pm 11257.029) + 27.040(\pm 73.678)T - 0.047(\pm 0.1205)T^2 \text{for (PNHA)} \quad (6)$$

where, V_ϕ^0 is the apparent molar volume at infinite dilution PBHA, PMNHA and PNHA, respectively and also, that the high values of parameter errors are due to the fitting in small degree of polynomial. That's why you have some small zigzags in Fig. 4.

The limiting apparent molar expansibilities are computed by using the relation:

$$\phi^0_E = (\partial V_\phi^0/\partial T)_P \quad (7)$$

The temperature derivative of limiting apparent molar expansibilities has been calculating using the following relation:

$$(\phi^0_E/\partial T) = (\partial^2 V_\phi^0/\partial T^2)_P \quad (8)$$

Table 3
Density, ρ, refractive index, n, apparent molar volume, V_ϕ, molar volume, V, refractivity, μ, specific refraction, R_S, molar refraction, R_M, polarizability, α, of hydroxamic acids in DMSO at different temperatures.

$m.10^{-2}$ (mol kg^{-1})	$P.10^{-3}$ (kg.m^{-3})	n	V_ϕ / . 10^{6} (m^3.mol^{-1})	V . 10^{-6} (m^3.mol^{-1})	μ	R_S .10^{-3} (kg.m^{-3})	R_M.10^{-6} (m^3.mol^{-1})	α . 10^{-3} (m^3.mol^{-1})
PBHA, T = 298.15 K								
0.0234	1.0966	1.4780	134.064	71.453	0.4780	0.2582	20.235	0.8025
0.0469	1.0981	1.4791	137.428	71.563	0.4791	0.2583	20.296	0.8049
0.0703	1.0991	1.4801	144.391	71.705	0.4801	0.2585	20.373	0.8080
0.0938	1.0999	1.4810	149.598	71.861	0.4810	0.2587	20.450	0.8110
0.1172	1.1009	1.4819	151.231	72.005	0.4819	0.2589	20.524	0.8139
0.1407	1.1019	1.4827	152.272	72.149	0.4827	0.2590	20.594	0.8167
0.1641	1.1033	1.4839	150.902	72.267	0.4839	0.2593	20.671	0.8198
0.1876	1.1041	1.4849	152.556	72.426	0.4849	0.2595	20.753	0.8230
0.2110	1.1050	1.4860	153.411	72.578	0.4860	0.2598	20.837	0.8264
0.2345	1.1060	1.4869	153.703	72.725	0.4869	0.2600	20.912	0.8293
PBHA, T = 303.15 K								
0.0234	1.0920	1.4771	116.428	71.755	0.4771	0.2588	20.278	0.8042
0.0469	1.0938	1.4782	123.391	71.846	0.4782	0.2589	20.344	0.8068
0.0703	1.0955	1.4791	126.772	71.943	0.4791	0.2589	20.404	0.8092
0.0938	1.0966	1.4798	133.782	72.080	0.4798	0.2589	20.467	0.8117
0.1172	1.0982	1.4807	134.300	72.184	0.4807	0.2590	20.529	0.8141
0.1407	1.0996	1.4816	135.789	72.302	0.4816	0.2591	20.596	0.8168
0.1641	1.1010	1.4828	136.801	72.421	0.4828	0.2593	20.675	0.8199
0.1876	1.1022	1.4841	138.428	72.553	0.4841	0.2596	20.763	0.8234
0.2110	1.1034	1.4853	139.656	72.686	0.4853	0.2599	20.843	0.8266
0.2345	1.1043	1.4861	141.708	72.840	0.4861	0.2600	20.916	0.8295
PBHA, T = 308.15 K								
0.0234	1.0878	1.4750	112.952	72.033	0.4750	0.2588	20.280	0.8043
0.0469	1.0900	1.4765	114.526	72.098	0.4765	0.2590	20.353	0.8072
0.0703	1.0922	1.4778	114.895	72.162	0.4778	0.2591	20.419	0.8098
0.0938	1.0942	1.4789	116.780	72.240	0.4789	0.2591	20.481	0.8122
0.1172	1.0956	1.4796	122.189	72.358	0.4796	0.2591	20.540	0.8146
0.1407	1.0969	1.4804	126.342	72.484	0.4804	0.2592	20.605	0.8172
0.1641	1.0983	1.4815	128.736	72.603	0.4815	0.2593	20.680	0.8201
0.1876	1.0996	1.4828	130.944	72.729	0.4828	0.2596	20.763	0.8234
0.2110	1.1008	1.4842	133.032	72.862	0.4842	0.2600	20.853	0.8270
0.2345	1.1018	1.4849	135.407	73.010	0.4849	0.2601	20.921	0.8297
PBHA, T = 313.15 K								
0.0234	1.0821	1.4738	94.810	72.414	0.4738	0.2596	20.343	0.8068
0.0469	1.0849	1.4747	94.565	72.438	0.4747	0.2594	20.383	0.8084
0.0703	1.0876	1.4757	95.542	72.470	0.4757	0.2592	20.429	0.8102
0.0938	1.0898	1.4768	100.485	72.535	0.4768	0.2592	20.488	0.8125
0.1172	1.0923	1.4780	101.160	72.580	0.478	0.2591	20.545	0.8148
0.1407	1.0943	1.4793	104.585	72.659	0.4793	0.2593	20.615	0.8175
0.1641	1.0960	1.4806	108.543	72.758	0.4806	0.2595	20.691	0.8205
0.1876	1.0975	1.4817	112.381	72.871	0.4817	0.2596	20.763	0.8234
0.2110	1.0995	1.4837	113.275	72.951	0.4837	0.2601	20.860	0.8272
0.2345	1.0999	1.4840	119.859	73.140	0.4840	0.2601	20.925	0.8298
$m.10^{-2}$ (mol kg^{-1})	$P.10^{-3}$ (kg.m^{-3})	n	V_ϕ / . 10^{6} (m^3.mol^{-1})	V . 10^{-6} (m^3.mol^{-1})	μ	R_S .10^{-3} (kg.m^{-3})	R_M.10^{-6} (m^3.mol^{-1})	α . 10^{-3} (m^3.mol^{-1})
PMNHA, T = 298.15 K								
0.0184	1.0967	1.4799	166.518	71.473	0.2590	0.4799	20.300	0.8050
0.0368	1.0984	1.4812	168.523	71.595	0.2592	0.4812	20.382	0.8083
0.0551	1.1000	1.4826	170.537	71.724	0.2594	0.4826	20.470	0.8117
0.0735	1.1011	1.4842	177.137	71.887	0.2599	0.4842	20.574	0.8158
0.0919	1.1024	1.4860	179.182	72.037	0.2604	0.4860	20.682	0.8202
0.1103	1.1037	1.4875	180.472	72.187	0.2608	0.4875	20.780	0.8241
0.1286	1.1048	1.4891	182.649	72.352	0.2613	0.4891	20.886	0.8283
0.1470	1.1059	1.4909	184.232	72.518	0.2619	0.4909	20.999	0.8328
0.1654	1.1072	1.4922	184.390	72.670	0.2622	0.4922	21.091	0.8366
0.1838	1.1082	1.4938	185.868	72.844	0.2627	0.4938	21.199	0.8410
PMNHA, T = 303.15 K								
0.0184	1.0921	1.4767	143.976	71.775	0.2586	0.4767	20.270	0.8038
0.0368	1.0935	1.4784	164.339	71.918	0.2591	0.4784	20.372	0.8080
0.0551	1.0952	1.4801	166.364	72.041	0.2595	0.4801	20.469	0.8120
0.0735	1.0969	1.4818	167.244	72.166	0.2599	0.4818	20.566	0.8160
0.0919	1.0981	1.4834	172.291	72.323	0.2603	0.4834	20.670	0.8199
0.1103	1.0996	1.4848	173.266	72.462	0.2606	0.4848	20.761	0.8235
0.1286	1.1011	1.4866	173.894	72.601	0.2611	0.4866	20.866	0.8277
0.1470	1.1026	1.4881	174.305	72.740	0.2614	0.4881	20.961	0.8316
0.1654	1.1041	1.4899	174.570	72.881	0.2619	0.4899	21.067	0.8356
0.1838	1.1055	1.4914	175.202	73.028	0.2621	0.4914	21.165	0.8392

(*continued on next page*)

Table 3 (*continued*)

PMNHA, $T = 308.15$ K								
0.0184	1.0880	1.4745	134.885	72.046	0.2585	0.4745	20.266	0.8037
0.0368	1.0897	1.4758	153.076	72.170	0.2588	0.4758	20.349	0.8071
0.0551	1.0916	1.4773	155.871	72.281	0.2590	0.4773	20.435	0.8104
0.0735	1.0934	1.4788	158.289	72.400	0.2593	0.4788	20.523	0.8139
0.0919	1.0950	1.4807	161.491	72.532	0.2598	0.4807	20.630	0.8181
0.1103	1.0968	1.4824	161.988	72.651	0.2601	0.4824	20.727	0.8220
0.1286	1.0981	1.4839	165.600	72.804	0.2605	0.4839	20.825	0.8259
0.1470	1.0997	1.4854	166.499	72.937	0.2608	0.4854	20.919	0.8297
0.1654	1.1014	1.4869	166.621	73.065	0.2612	0.4869	21.010	0.8335
0.1838	1.1030	1.4889	167.137	73.199	0.2616	0.4889	21.123	0.8377
PMNHA, $T = 313.15$ K								
0.0184	1.0821	1.4730	120.977	72.440	0.2593	0.4730	20.322	0.8060
0.0368	1.0846	1.4743	127.671	72.512	0.2592	0.4743	20.390	0.8084
0.0551	1.0872	1.4755	128.139	72.577	0.2591	0.4755	20.452	0.8109
0.0735	1.0896	1.4769	130.556	72.655	0.2593	0.4769	20.526	0.8140
0.0919	1.0916	1.4784	135.628	72.761	0.2594	0.4784	20.611	0.8171
0.1103	1.0939	1.4801	136.572	72.847	0.2597	0.4801	20.698	0.8207
0.1286	1.0955	1.4817	141.851	72.981	0.2600	0.4817	20.795	0.8243
0.1470	1.0975	1.4832	143.398	73.087	0.2603	0.4832	20.881	0.8281
0.1654	1.0990	1.4848	147.152	73.229	0.2607	0.4848	20.980	0.8320
0.1838	1.1001	1.4868	151.996	73.398	0.2613	0.4868	21.103	0.8369
$m.10^{-2}$ (mol kg^{-1})	$P.10^{-3}$ (kg.m^{-3})	n	V_ϕ / . 10^{6} (m^3.mol^{-1})	V . 10^{-6} (m^3.mol^{-1})	μ	R_S .10^{-3} (kg.m^{-3})	R_M.10^{-6} (m^3.mol^{-1})	α . 10^{-3} (m^3.mol^{-1})
PNHA, $T = 298.15$ K								
0.0234	1.0970	1.4791	145.099	71.448	0.4791	0.2585	20.264	0.8036
0.0469	1.0990	1.4804	146.981	71.545	0.4804	0.2587	20.339	0.8066
0.0703	1.1009	1.4814	148.869	71.650	0.4814	0.2587	20.405	0.8092
0.0938	1.1027	1.4827	150.765	71.761	0.4827	0.2589	20.484	0.8123
0.1172	1.1045	1.4838	151.801	71.872	0.4838	0.2589	20.555	0.8152
0.1407	1.1062	1.4850	153.131	71.990	0.4850	0.2591	20.633	0.8182
0.1641	1.1076	1.4862	155.878	72.129	0.4862	0.2593	20.716	0.8215
0.1876	1.1089	1.4871	158.430	72.275	0.4871	0.2594	20.791	0.8245
0.2110	1.1100	1.4880	161.342	72.435	0.4880	0.2596	20.870	0.8276
0.2345	1.1110	1.4893	164.074	72.602	0.4893	0.2599	20.965	0.8314
PNHA, $T = 303.15$ K								
0.0234	1.0921	1.4778	136.647	71.770	0.4778	0.2591	20.308	0.8054
0.0469	1.0943	1.4790	138.538	71.855	0.4790	0.2591	20.376	0.8081
0.0703	1.0965	1.4804	138.980	71.940	0.4804	0.2593	20.451	0.8110
0.0938	1.0985	1.4817	141.244	72.039	0.4817	0.2594	20.527	0.8140
0.1172	1.0998	1.4826	148.617	72.184	0.4826	0.2595	20.601	0.8170
0.1407	1.1018	1.4837	148.348	72.283	0.4837	0.2595	20.669	0.8197
0.1641	1.1032	1.4849	151.841	72.423	0.4849	0.2597	20.753	0.8230
0.1876	1.1047	1.4858	153.857	72.557	0.4858	0.2598	20.824	0.8258
0.2110	1.1058	1.4868	157.335	72.718	0.4868	0.2600	20.907	0.8291
0.2345	1.1073	1.4884	158.310	72.853	0.4884	0.2604	21.005	0.8330
PNHA, $T = 308.15$ K								
0.0234	1.0879	1.4759	132.404	72.048	0.4759	0.2592	20.318	0.8057
0.0469	1.0899	1.4770	140.892	72.147	0.4770	0.2593	20.386	0.8084
0.0703	1.0919	1.4785	143.539	72.246	0.4785	0.2595	20.469	0.8117
0.0938	1.0938	1.4798	145.827	72.352	0.4798	0.2596	20.546	0.8148
0.1172	1.0953	1.4806	150.623	72.486	0.4806	0.2596	20.614	0.8175
0.1407	1.0969	1.4818	153.005	72.613	0.4818	0.2598	20.694	0.8207
0.1641	1.0985	1.4831	154.639	72.740	0.4831	0.2600	20.778	0.8240
0.1876	1.1002	1.4840	155.249	72.861	0.4840	0.2600	20.846	0.8267
0.2110	1.1011	1.4850	159.623	73.038	0.4850	0.2603	20.933	0.8301
0.2345	1.1026	1.4863	160.413	73.174	0.4863	0.2605	21.020	0.8336
PNHA, $T = 313.15$ K								
0.0234	1.0819	1.4741	123.686	72.449	0.4741	0.2598	20.365	0.8076
0.0469	1.0843	1.4754	127.826	72.522	0.4754	0.2598	20.433	0.8103
0.0703	1.0868	1.4772	127.532	72.588	0.4772	0.2601	20.518	0.8137
0.0938	1.0896	1.4786	123.910	72.635	0.4786	0.2601	20.583	0.8162
0.1172	1.0915	1.4799	129.613	72.742	0.4799	0.2602	20.661	0.8193
0.1407	1.0929	1.4809	137.036	72.884	0.4809	0.2603	20.738	0.8224
0.1641	1.0944	1.4821	141.637	73.019	0.4821	0.2605	20.821	0.8257
0.1876	1.0958	1.4829	145.589	73.162	0.4829	0.2606	20.891	0.8285
0.2110	1.0972	1.4841	148.614	73.305	0.4841	0.2608	20.976	0.8319
0.2345	1.0983	1.4854	152.338	73.469	0.4854	0.2611	21.071	0.8356

m is the Molarities of hydroxamic acid in the solvent (DMSO). Standard uncertainties μ are μ (m) = 0.0001, u (ρ) 2.10^{-4} kg.m^{-3}, u (n) =0.0001, u (V_ϕ) = 0.003 m^3.mol^{-1}, u (V) = 0.001 m^3. mol^{-1}, u (μ) =0.0001, u (R_S) = 0.0001cm^{-3}, u (R_M) = 0.006 m^3. mol^{-1}, u (α) = 0.0003 cm^3. mol^{-1} .

Table 4
Apparent molar volume at infinite dilution, V_ϕ^0, experimental slop, S_V^*, and apparent molar expansibilities at infinite dilution, ϕ^{0}_{E}, thermal expansion coefficient, α_2, Hepler's constant, $\partial^2 V_\phi^0/\partial T^2$, of hydroxamic acids in DMSO at different temperatures.

T/K	V_ϕ^0 . 10^6 (m^3. mol^{-1})	S_V^* .10^6 (m^3. $mol^{-3/2}$.$l^{1/2}$)	ϕ^{0}_{E} . 10^6(m^3. mol^{-1})	α_2 .10^6 (K^{-1})	$\partial^2 V_\phi^0/\partial T^2$ .10^6 (m^3. mol^{-1})
PBHA					
298.15	127.277(±2.98)	60.105(±8.31)	0.6494	0.0051	
303.15	107.895(±2.06)	72.119(±5.76)	0.6294	0.0058	-0.0040
308.15	97.818(±2.54)	74.880(±7.06)	0.6094	0.0062	
313.15	78.223(±3.32)	76.435(±9.26)	0.5894	0.0075	
PMNHA					
298.15	155.945(±1.53)	72.246(±4.82)	-1.3984	-0.0089	
303.15	142.380(±5.43)	85.901(±17.0)	-2.4784	-0.0174	-0.2160
308.15	130.526(±4.49)	93.962(±14.1)	-3.5584	-0.0272	
313.15	106.127(±2.30)	99.369(±7.24)	-4.6384	-0.0437	
PNHA					
298.15	134.587(±1.88)	60.934(±5.76)	-0.9861	-0.0073	
303.15	122.598(±2.31)	79.240(±7.09)	-1.4561	-0.0118	-0.0940
308.15	121.690(±1.32)	89.342(±4.06)	-1.9261	-0.0158	
313.15	104.913(±5.15)	98.724(±15.8)	-2.3961	-0.0228	

Standard uncertainties μ are μ (T)=0.01K, u (V_ϕ^0) 0.003 m^3. mol^{-1}, u (S_V^*) = 0.0004 m^3. $mol^{-3/2}$.$l^{1/2}$, u (ϕ^{0}_{E}) =0.0006 m^3. mol^{-1}, u (α_2) =0.0001 K^{-1}, u ($\partial^2 V_\phi^0/\partial T^2$)=0.0001 m^3. mol^{-1}.

Table 5
Excess molar volume, V^E, refractive index deviation, n^E, and molar refraction deviation, $R^E_{M,}$ of hydroxamic acids (PBHA, PMNHA and PNHA)) in DMSO (1) at different temperatures.

Mole fraction, x_2	V^E .10^{-6} (m^3 mol^{-1})				n^E				R^E_M 10^6 (m^3 • mol^{-1})		
	298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
PBHA												
0.0017	-0.1107	-0.1446	-0.1524	-0.1876	0.0010	0.0018	0.0020	0.0028	0.0140	0.0247	0.0302	0.0504
0.0034	-0.2087	-0.2632	-0.2985	-0.3754	0.0021	0.0029	0.0035	0.0037	0.0172	0.0314	0.0439	0.0308
0.0050	-0.2744	-0.3756	-0.4448	-0.5566	0.0031	0.0038	0.0048	0.0047	0.0351	0.0326	0.0503	0.0168
0.0067	-0.3273	-0.4488	-0.5781	-0.7047	0.0040	0.0045	0.0059	0.0058	0.0531	0.0359	0.0531	0.0157
0.0084	-0.3935	-0.5552	-0.6722	-0.8730	0.0049	0.0054	0.0066	0.0070	0.0675	0.0389	0.0525	0.0127
0.0102	-0.4600	-0.6487	-0.7599	-1.0083	0.0057	0.0063	0.0074	0.0083	0.0782	0.0457	0.0574	0.0227
0.0119	-0.5530	-0.7426	-0.8546	-1.1240	0.0069	0.0075	0.0085	0.0096	0.0961	0.0653	0.0714	0.0383
0.0136	-0.6069	-0.8236	-0.9430	-1.2268	0.0079	0.0088	0.0098	0.0107	0.1179	0.0924	0.0947	0.0504
0.0153	-0.6676	-0.9049	-1.0250	-1.3631	0.0090	0.0100	0.0112	0.0127	0.1416	0.1121	0.1236	0.0861
0.0171	-0.7352	-0.9667	-1.0942	-1.3935	0.0099	0.0108	0.0119	0.0130	0.1562	0.1248	0.1305	0.0897
PMNHA												
0.0013	-0.1172	-0.1511	-0.1655	-0.1874	0.0029	0.0014	0.0015	0.0020	0.0718	0.0083	0.0082	0.0209
0.0026	-0.2283	-0.2434	-0.2784	-0.3551	0.0042	0.0031	0.0028	0.0033	0.0876	0.0443	0.0240	0.0217
0.0040	-0.3332	-0.3559	-0.4049	-0.5297	0.0056	0.0048	0.0043	0.0045	0.1089	0.0747	0.0434	0.0169
0.0053	-0.4059	-0.4687	-0.5252	-0.6914	0.0072	0.0065	0.0058	0.0059	0.1468	0.1051	0.0647	0.0231
0.0066	-0.4919	-0.5490	-0.6326	-0.8267	0.0090	0.0081	0.0077	0.0074	0.1883	0.1413	0.1044	0.0406
0.0080	-0.5784	-0.6494	-0.7537	-0.9824	0.0105	0.0095	0.0094	0.0091	0.2189	0.1647	0.1330	0.0597
0.0093	-0.6521	-0.7502	-0.8421	-1.0919	0.0121	0.0113	0.0109	0.0107	0.2571	0.2027	0.1638	0.0884
0.0107	-0.7261	-0.8514	-0.9507	-1.2285	0.0139	0.0128	0.0124	0.0122	0.3027	0.2298	0.1889	0.1058
0.0120	-0.8137	-0.9530	-1.0663	-1.3321	0.0152	0.0146	0.0139	0.0138	0.3264	0.2679	0.2122	0.1364
0.0134	-0.8818	-1.0484	-1.1757	-1.4095	0.0168	0.0161	0.0159	0.0158	0.3668	0.2970	0.2558	0.1895
PNHA												
0.0014	-0.1369	-0.1513	-0.1591	-0.1743	0.0021	0.0025	0.0029	0.0031	0.0372	0.0482	0.0613	0.0651
0.0028	-0.2677	-0.2965	-0.2921	-0.3356	0.0034	0.0037	0.0040	0.0044	0.0473	0.0509	0.0642	0.0677
0.0042	-0.3923	-0.4419	-0.4254	-0.5039	0.0044	0.0051	0.0055	0.0062	0.0484	0.0609	0.0817	0.0867
0.0056	-0.5107	-0.5746	-0.5525	-0.6924	0.0057	0.0064	0.0068	0.0076	0.0622	0.0709	0.0938	0.0853
0.0070	-0.6296	-0.6618	-0.6535	-0.8213	0.0068	0.0073	0.0076	0.0089	0.0688	0.0795	0.0951	0.0972
0.0084	-0.7423	-0.7952	-0.7615	-0.9173	0.0080	0.0084	0.0088	0.0099	0.0808	0.0823	0.1091	0.1075
0.0098	-0.8358	-0.8897	-0.8699	-1.0203	0.0092	0.0096	0.0101	0.0111	0.0984	0.0999	0.1269	0.1233
0.0112	-0.9232	-0.9910	-0.9852	-1.1170	0.0101	0.0105	0.0110	0.0119	0.1071	0.1047	0.1280	0.1263
0.0126	-0.9979	-1.0665	-1.0480	-1.2141	0.0110	0.0115	0.0120	0.0131	0.1195	0.1208	0.1480	0.1441
0.0141	-1.0664	-1.1687	-1.1509	-1.2915	0.0123	0.0131	0.0133	0.0144	0.1484	0.1513	0.1677	0.1713

Standard uncertainties μ are μ (x_2)=0.0001, u (V^E) 0.0003 m^3. mol^{-1}, u (n^E) = 0.0002, u (R^E_M) =0.0003 m^3. mol^{-1}.

where, ϕ^{0}_{E} is apparent molar expansibilities at infinite dilution and p is the pressure. The ϕ^{0}_{E} values are an important indicator of solute-solvent interactions. The data of ϕ^{0}_{E} of the solutes in the solvent can be useful in interpreting the structure making or breaking properties of the solutes. It is evident from Table 4 that the values of ϕ^{0}_{E} are negative (i.e., decreasing volume with increasing temperature) which show highly hydrophobic characters of these molecules. From the apparent molar volume at infinite dilution V_ϕ^0, the values of the thermal expansion coefficients of the solute at infinite dilution α_2 are also determined using the following equation [40] and are given in Table 4:

$$\alpha_2 = \phi^{0}_{E}/V_\phi^0 \quad (9)$$

Furthermore, the values of thermal expansion coefficient, α_2, as in Table 4 show a decrease with increasing temperature. When the temperature is increased, the density of the solution decreases, Table 3, resulting in the thermal expansivity coefficient, α_2.

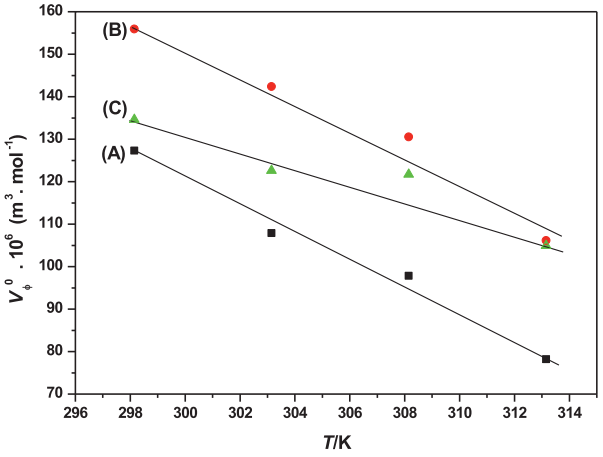


Fig. 4. Plot of apparent molar volume at infinite dilution (V_ϕ^0) vs T=(298.15, 303.15 308.15 and 313.15) K of (■) PBHA, (●) PMNHA and (►) PNHA in DMSO.

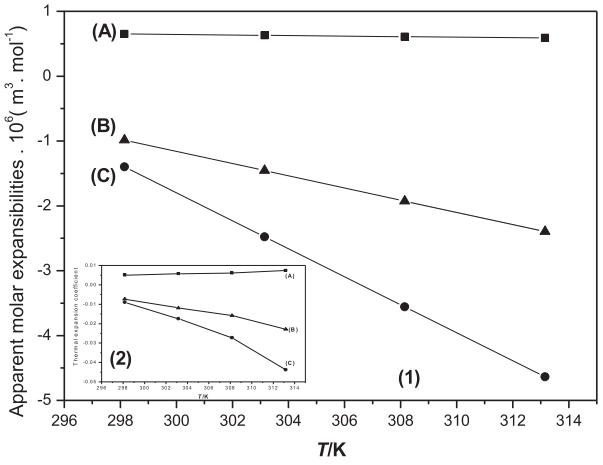


Fig. 5. Plot of (1) apparent molar expansibilities at infinite dilution (ϕ^0_E), **and (2)** thermal expansion coefficient, (α_2) vs T=(298.15, 303.15 308.15 and 313.15) K of (■) PBHA, (●) PMNHA and (►) PNHA in DMSO.

The Hepler's constant, $\partial^2 V_\phi^0/\partial T^2$, [41] has been calculated using Eq. (3) and the values are listed in Table 4. According to Hepler, the sign of $\partial^2 V_\phi^0/\partial T^2$ is a better criterion in characterizing the structure making and breaking ability of the solutes in solutions. If $\partial^2 V_\phi^0/\partial T^2$ is positive, the solute is structure maker and it has a negative value for a structure breaker solute. In the present investigation, the observed positive values of Hepler's constant, $\partial^2 V_\phi^0/\partial T^2$, for three molecules suggest the structure promoter nature of these solutes in solution.

The molar volume, V, of the solutions has been computed from the measured values of ρ, using the equation [42]:

$$V = \left(x_1 M_1 + x_2 M_2\right)/\rho \tag{10}$$

where, x_1, M_1 and x_2, M_2 are the mole fraction and molecular weight of the solvent and solute, respectively. The data are listed in Table 3.

3.2. Steric properties

The refractive indices, n, data have been used to calculate refractivity μ (= n-1) and specific refraction, R_s, of these reagents which are presented in Table 3. The values of the R_s are also computed as a function of their concentration in DMSO following the equation, proposed by Lorentz and Lorenz:

$$R_s = \left(n^2 - 1\right)/\left(n^2 + 2\right) \times 1/\rho \tag{11}$$

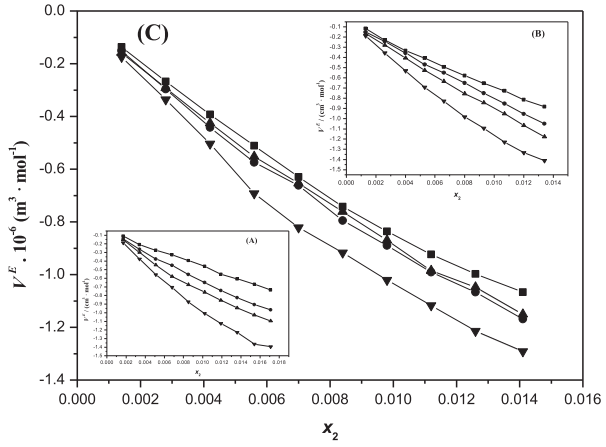


Fig. 6. Plot of excess molar volume (V^E) vs mole fraction (x_2) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures: ■, 298.15 K; ●, 303.15 K; ◄, 308.15 K; and ►, 313.15 K.

The n of a material is estimated in terms of the molar refraction, R_M, which quantifies the intrinsic refractive power of the structural units constituting that material. A definition proposed for R_M that incorporates both of the key physical factors determining the n, is the molar refraction according to Lorentz and Lorentz [43]:

$$R_M = \left[\left(n^2 - 1/n^2 + 2\right)\right].V \tag{12}$$

where, V is the molar volume. The values of R_M are reported in Table 3. Values of R_M increase linearly with concentration and are proportional to the dispersive forces. Thus, the increasing magnitude of R_M for PBHA, PMNHA and PNHA in DMSO indicates strong solute-solvent interactions.

The n related to the polarizability, α, depends on geometry of the molecules, α is a fundamental molecular property of great importance, it defines the nature of intermolecular attractive force for non-polar molecules. Therefore, in order to gain further information about specific intermolecular interactions of any kind, α of the system is computed by equation:

$$\alpha = 3R_M/4\pi N \tag{13}$$

where, α is the electronic polarizability and N is Avogadro's number. α of molecules is one of the most significant electric property, which characterizes the ability of the electronic system to be distorted by the external electric filed. Table 3 enlists the values of α and shows that α, of hydroxamic acid solutions increases with concentration. This trend is slightly influenced by temperature and the obvious decrease with temperature indicates the presence of intermolecular and intramolecular interactions between the molecules of the solute and the solvent. n characteristics show that the dipole in the compound lies perpendicular to the longer axis of the molecules and is responsible for the intermolecular attractions which is further accompanied by increase in the value of R_M and α with increasing concentration of solution because of mutual compensation of dipoles.

3.3. Excess properties

The excess properties of the solution are calculated using the following equation:

$$Y^E = Y - \left(x_1 Y_1 + x_2 Y_2\right) \tag{14}$$

where, Y^E represents the excess molar volume, V^E, or refractive index deviation, n^E, or molar refraction deviation, R_M^E . The V^E, n^E and R_M^E of DMSO, PBHA, PMNHA, PNHA and their solutions are denoted by Y_1, Y_2 and Y, respectively. x_1 and x_2, are mole fraction of solvent and solute, respectively. The calculated values of V^E, n^E and R_M^E are listed in Table

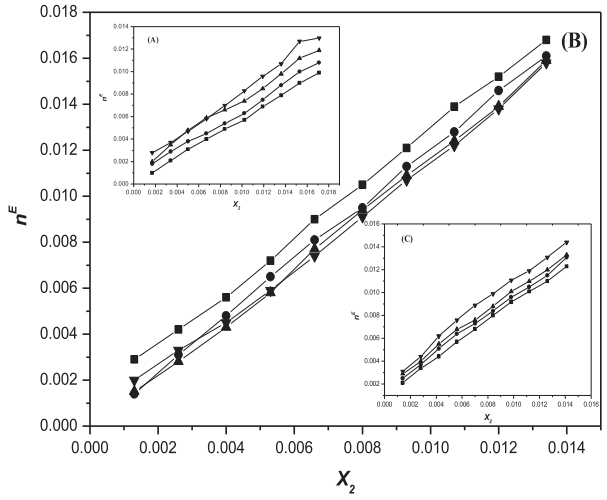


Fig. 7. Plot of refractive index deviation (n^E) vs mole fraction (x_2) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures: ■, 298.15 K; ●, 303.15 K; ◄, 308.15 K; and ►, 313.15 K.

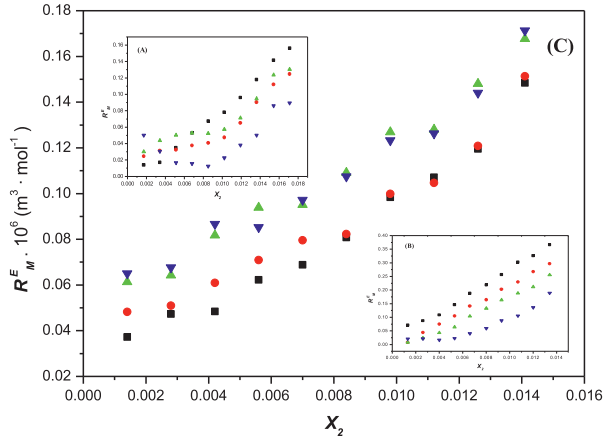


Fig. 8. Plot of molar refraction deviation (R^E_M) vs mole fraction (x_2) of (A) PBHA, (B) PMNHA and (C) PNHA in DMSO at different temperatures: ■, 298.15 K; ●, 303.15 K; ◄, 308.15 K; and ►, 313.15 K.

5. According to Nakata and Sakurai [44] the sign of n^E is opposite to that of V^E, if the behavior of n is not linear between n_1 and n_2. This rule is truly fulfilled for all the hydroxamic acids. The values of n^E are positive over the entire concentration range and at different temperatures. Thus, dispersion forces in the solution are higher than in solvent. The knowledge of V^E help in understanding the molecular orientation and to study the extent of intermolecular interactions between solvent molecules of the solution. V^E is influenced by: (i) physical interactions mainly due to dispersive force, (ii) the dipole-dipole and donor-acceptor interaction between unlike molecules and (iii) the filling of smaller molecules into the voids created by bigger molecules. V^E is a measure of the deviations from the actual property if the system behaves ideally and gives information on molecular interactions between the solvent molecules of the solution and are influenced by effects such as differences in shape and size of solvent molecules, reorientation of the solvent molecules in the solution, and intermolecular interactions [45,46]. It is well known that V^E is the result of several opposing effects. Interaction between like molecules lead to increased V^E values, while negative contribution arises from interactions between unlike molecules, or structural effects as change in free volume or interstitial accommodation. For the investigated systems, the negative V^E values may be due to interactions between unlike molecules as shown in Fig. 6.

The refractive index deviation, n^E, is positive over the whole concentration range and at various temperatures. As indicated in Fig. 7, the value increases with temperature. The knowledge of V^E helps in understanding the molecular orientation and to study the extent of intermolecular interactions between solvent molecules of the solution. The negative values of R^E_M throughout the concentration and temperature range, as shown in the Fig. 8, indicate the presence of strong intermolecular interactions.

We can remark that the effect of temperature is not very significant for the hydroxamic acids due to the smaller n^E and V^E values with increase in temperature.

4. Conclusions

The hydroxamic acid functional group –NOH•C=O shows a wide range of biological activity. Using density data, partial molar volumes, apparent molar expansibilities at infinite dilution, and molar refraction, the excess properties have been computed. The behavior of the parameters suggests strong solute-solvent interaction. The thermal expansion coefficients of the three molecules are also determined. The negative V^E valuesand positive $R^E m$ values suggest that PBHA, PMNHA, and PNHA acts as structure maker in DMSO though hydrogen bond formation. As hydroxamic acids are bioactive molecules these parameters will be of further use in Quantitative Structuer Activity Relationship studies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interestsor personal relationships that could have appeared to influence the work reported in this paper.

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