

Study of Molecular Interaction in Binary Mixtures of *N,N*-Dimethylformamide with 2-Butanol and 2-Pentanol at Temperatures Between 298.15 K and 323.15 K

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ABSTRACT

A study of viscosity for the binary mixtures of *N,N*-dimethylformamide with 2-butanol and 2-pentanol at atmospheric pressure and at temperatures between 298.15 K and 323.15 K have been measured over the entire range of composition. From these data, enthalpy of activation (ΔH^\ddagger), excess enthalpy of activation ($\Delta H^{\ddagger E}$), entropy of activation (ΔS^\ddagger), excess entropy of activation ($\Delta S^{\ddagger E}$) and interaction parameter (ϵ) have been calculated. The Redlich-Kister polynomial equation was used for correlating all the results. The obtained data were compared with those available in literature. The behavior of alcohols and interactions between the components mixture was performed using different excess thermodynamic properties. A discussion of molecular interaction has been done between the components of the mixture.

Keywords: Molecular interaction; Viscosity; Interaction parameter; Entropy of activation; 2-Butanol; 2-Pentanol; Redlich-Kister polynomial equation.

INTRODUCTION

The viscosity (η) measurements find wide applications in characterizing the physico-chemical behavior of liquid mixtures and in the study of molecular interactions in pure liquids and binary mixtures. Such data are used subsequently for research as well as industrial purposes. Among the physical properties of fluids needed to optimize the industrial process design, viscosity is one of the most important. In chemical industry, viscosity is necessary for hydraulic calculations, fluid transport through pipes and pore surfaces, and in many mass and

energy transfers.¹ The thermodynamic properties of binary liquid mixtures containing protic, aprotic, and associated liquids have been studied previously.²⁻⁶ Such properties provide insight about the state of liquids in pure as well as the mixed state along with the interactions operating among the constituent molecules and can also be used for the development or testing of theories or models describing the thermodynamic behavior of mixtures.⁷⁻⁹ The thermodynamic properties of liquid mixtures in which the molecular interactions are mainly due to association through hydrogen bonding between carbonyl and hydroxyl groups have been studied earlier¹⁰⁻¹²

The molecular interaction is associated with the alkanol hybrids, which are one of the most unique components as the alkanols group is so much polar, because of the deepness with other groups which is the attraction of the polar ends.¹³ The binary liquid mixtures that are polar consists of one associative and other non-associative liquids, which provide much valuable information regarding the molecular interaction in solutions.¹⁻³ Among these kinds of associative liquids, the alcohols are the center of interest to scientists. Hydrogen bonding in liquid systems has been intensively studied for a long time and it is still subject to a lively scientific debate because of its outstanding role in chemical arena.¹⁴

The amides containing multi-component solvent systems are interesting liquid systems for the study of molecular interactions. Because in chemical reactions and in many industrial processes the amides are the most common solvents used. Also, for the investigation of peptide and protein interactions in biological systems, amides are the convenient model systems.^{15,16}

Molecular interactions of *N,N*-dimethylformamide (DMF) with some solvents reported by various thermodynamic and thermophysical measurements.¹⁷ DMF is aprotic and unassociated in its pure liquid state.¹⁸ Molecular interactions of DMF with some solvents reported by various thermodynamic and thermophysical measurements. Several topics and examples of thermodynamic studies are depicted on the basis of the structural behavior of DMF for binary and ternary mixtures of non-electrolytes.¹⁹ Solutions of DMF are used to process polymer fibers, films, and surface coatings; to permit easy spinning of acrylic fibers, also to produce wire enamels, and as a crystallization medium in the pharmaceutical industry.

It is possible to determine the interesting properties by the binary mixtures of different alkanols due to their complexity, specific interaction, and hydrogen bond effects.²⁰⁻²³ Also, alcohols and ethers containing mixtures seem to be very interesting due to their increasingly used as additives to gasoline owing to their octane-enhancing and pollution reducing properties.²⁴ and these kinds of mixtures show a particular challenge in order to express the interaction behavior between the solutions.²⁵

Alkanols of short chain length have a greater proton-donor capacity, so the strength of bonding is expected to decrease with an increase in their chain lengths. Moreover, because of the relevant steric hindrance of alkyl groups, hydrogen bonds are weakened for higher alkanols,^{26,27} and hence, they influence physical properties of the alkanols correspondingly. Alkanols are the -OH functional group containing compounds bonded to a carbon atom. These polar liquids are self-associated through hydrogen bonding, creating multimers of different degrees.²⁸⁻³⁰ This kind of association is disturbed when they are mixed with another solvent and when amide and alkanols are mixing, different intermolecular interactions take place, resulting in non-ideal behavior.³¹

EXPERIMENTAL SECTION

Materials and Method

2-BuOH (Aldrich, purity 99+%), 2-PnOH (Aldrich, purity 98%) and DMF (Aldrich, purity HPLC grade 99.9+ %) were used without further treatment as shown in Figure 1.

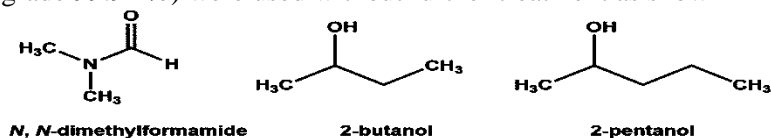


Figure 1: Structures of the compounds used

The viscosities of pure chemicals were compared with literature values, which cover satisfactory agreements as shown in Table 1. All of the measurements on viscosities (η) were carried out on an Anton Paar, SVM 3000 (Austria-Europe) rotational Stabinger viscometer. The mixtures samples were prepared by mixing the pure components at different proportions up to ± 0.0001 g, which was then converted into mole fraction. All the molar quantities used here are based on the IUPAC relative atomic mass table.³²

Table 1: Experimental (bold letter) and literature values of viscosities, η /mPa.s of the pure *N,N*-dimethylformamide (DMF), 2-butanol (2-BuOH) and 2-pentanol (2-PnOH) at temperatures 298.15 K - 323.15 K.

Temp. T / K	2-BuOH	2-PnOH	DMF
298.15	2.9575 2.996 ³³ 2.998 ³⁵	3.3320 3.478 ³⁴ 3.47 ³⁶	0.79780 0.799 ⁴⁸ 0.80 ⁵¹
303.15	2.4604 2.492 ³⁹ 2.495 ⁴⁰	2.7433 2.77 ⁴³ 2.7385 ⁵³ 2.882 ⁴⁴ 2.6985 ⁵⁴	0.75038 0.766 ⁴⁹ 0.7560 ⁵² 0.7485 ⁵⁰
308.15	2.0620 2.0042 ³⁷ 2.011 ³⁸ 2.0542 ⁴⁵	2.2799 2.3120 ⁵³	0.70699 0.711 ⁴⁹ 0.7063 ⁵⁰ 0.7262 ⁵⁵
313.15	1.7402 1.7833 ⁴¹ 1.785 ⁴²	1.9121 1.9429 ⁵³	0.66716 0.6683 ⁴⁸ 0.6797 ⁵³ 0.6683 ⁵⁰ 0.684 ⁴⁷
318.15	1.4781 1.557 ⁴⁶ 1.533 ⁴⁷	1.6175 1.6606 ⁵³	0.63057
323.15	1.2666 1.3149 ⁴¹ 1.315 ⁴²	1.3863 1.4795 ⁵³	0.59613 0.6004 ⁵⁰ 0.617 ⁴⁹

RESULTS AND DISCUSSION

Viscosity and free energy

The measured data of viscosity, η , for the binary mixtures of 2-BuOH and 2-PnOH with DMF as the common component at different compositions and temperatures are calculated. The experimental viscosities, η , were fitted to the polynomial equation of the following form:

$$\eta = \sum_{i=0}^n a_i (1 - x_1)^i \quad (1)$$

Here, a_i is the i^{th} fitting coefficient and x_1 is the mole fraction of DMF. Eyring and his co-workers using absolute reaction rate theory and partition functions, correlated viscosity to η , as follows:

$$\eta = (hN/V_m) \cdot \exp(\Delta G^\ddagger)/RT \quad (2)$$

Where, ΔG^\ddagger is the free energy of activation per mole for viscous flow, h is the Planck's constant ($=6.6262 \times 10^{-34}$ J-sec), N is the Avogadro number ($=6.023 \times 10^{23}$ mol $^{-1}$), V_m is the molar volume for pure liquids or mixture, R is the molar gas constant ($=8.3145$ JK $^{-1}$ mol $^{-1}$), T is the absolute temperature in Kelvin scale and η is the observed viscosity in kgm $^{-1}$ s $^{-1}$.

Enthalpy of activation and entropy of activation

According to the definition of ΔG^\ddagger eq. (2) reduces to-

$$\eta = (hN/V_m) \cdot \exp(\Delta H^\ddagger - T\Delta S^\ddagger)/RT$$

$$\text{Or, } \ln(\eta V_m/hN) = -\Delta S^\ddagger/R + \Delta H^\ddagger/RT \quad (3)$$

Where, ΔH^\ddagger is the enthalpy of activation per mole and ΔS^\ddagger the entropy of activation per mole for viscous flow.

The plot of $\ln\left(\frac{\eta V_m}{hN}\right)$ vs. $1/T$ of eq. (3) gives a straight line with slope, $\Delta H^\ddagger/R$ and intercept, $-\Delta S^\ddagger/R$ assuming that ΔH^\ddagger and ΔS^\ddagger be almost independent of temperature. Therefore, ΔH^\ddagger and ΔS^\ddagger can easily be calculated from the slope and intercept of eq. (3) as,

$$\Delta H^\ddagger = \text{Slope} \times R \quad (4)$$

$$\Delta S^\ddagger = -\text{Intercept} \times R \quad (5)$$

Excess enthalpy of activation and excess entropy of activation

Excess enthalpy of activation ($\Delta H^{\ddagger E}$) and excess entropy of activation ($\Delta S^{\ddagger E}$) for viscous flow were calculated by using the following relations,

$$\Delta H^{\ddagger E} = \Delta H^\ddagger - \Delta H_{id}^\ddagger$$

$$\therefore \Delta H^{\ddagger E} = \Delta H^\ddagger - (x_1\Delta H_1^\ddagger + x_2\Delta H_2^\ddagger) \quad (6)$$

$$\text{And } \Delta S^{\ddagger E} = \Delta S^\ddagger - \Delta S_{id}^\ddagger$$

$$\therefore \Delta S^{\ddagger E} = \Delta S^\ddagger - (x_1\Delta S_1^\ddagger + x_2\Delta S_2^\ddagger) \quad (7)$$

Where, the subscripts 1 and 2 represent the pure components of the mixture. The excess thermodynamic parameters (Y^E), such as, $\Delta H^{\#E}$ and $\Delta S^{\#E}$ were also calculated by following a general equation of the form,

$$Y^E = Y - \sum x_i Y_i \quad (8)$$

Here, Y is the observed property of the mixture and Y_i is that for the i^{th} component.

The thermodynamic parameters, enthalpy ($\Delta H^{\#}$) and entropy ($\Delta S^{\#}$) of activation for viscous flow for the systems of 2-BuOH + DMF and 2-PnOH+ DMF have been estimated in composition range, $0 \leq x_1 \leq 1$ from the measured η and ρ data between 298.15 and 323.15 K.

All the values obtained for $\Delta H^{\#E}$ and $\Delta S^{\#E}$ are represented in Tables 2 & 3 and Figures 2-5 which shown the comparative diagrams of $\Delta H^{\#E}$ vs. x_1 and $\Delta S^{\#E}$ vs. x_1 , respectively. The Redlich-Kister coefficients for $\Delta H^{\#}$ and $\Delta S^{\#}$ relevant to standard deviation (σ) were also computed.⁵⁶

Table 2: Experimental enthalpy ($\Delta H^{\#}$), entropy ($\Delta S^{\#}$), excess enthalpy ($\Delta H^{\#E}$), excess entropy ($\Delta S^{\#E}$) of the systems DMF (x_1) + 2-BuOH (x_2) for different molar ratios.

x_1	$\Delta H^{\#}$ (kJ mol ⁻¹)	$\Delta H^{\#E}$ (kJ mol ⁻¹)	$\Delta S^{\#}$ (J mol ⁻¹ K ⁻¹)	$\Delta S^{\#E}$ (J mol ⁻¹ K ⁻¹)
0.0000	26.3078	0.0000	53.1300	0.0000
0.0506	23.1753	-2.2406	44.1477	-6.6078
0.0997	20.8641	-3.6667	37.6926	-10.7073
0.1515	18.2492	-5.3984	30.2122	-15.8368
0.2000	17.0905	-5.6810	27.2634	-16.4537
0.2497	16.4769	-5.3544	25.9837	-15.2309
0.2909	16.0505	-4.9261	25.1868	-13.7528
0.3510	15.2608	-4.8308	23.1572	-13.4268
0.3999	14.3976	-4.7912	20.8073	-13.3738
0.4494	13.4862	-4.8176	18.2987	-13.5268
0.5003	12.7222	-4.6787	16.2173	-13.2052
0.5491	12.0082	-4.5095	14.2211	-12.8506
0.6002	11.9216	-3.7040	14.2244	-10.4727
0.6500	11.6826	-3.0081	13.7906	-8.4182
0.7000	11.2285	-2.6128	12.6261	-7.3219
0.7499	10.3219	-2.6379	9.9862	-7.6157
0.7999	9.8897	-2.1745	8.8842	-6.3337
0.8500	9.7143	-1.4542	8.6605	-4.1734
0.9000	9.2225	-1.0592	7.4096	-3.0640
0.9500	8.9500	-0.4396	6.8388	-1.2603
1.0000	8.5010	0.0000	5.7340	0.0000

Table 3: Experimental enthalpy (ΔH^\ddagger), entropy (ΔS^\ddagger), excess enthalpy ($\Delta H^{\ddagger E}$), excess entropy ($\Delta S^{\ddagger E}$) of the systems DMF (x_1) + 2-PnOH (x_2) for different molar ratios.

x_1	ΔH^\ddagger (kJ mol ⁻¹)	$\Delta H^{\ddagger E}$ (kJ mol ⁻¹)	ΔS^\ddagger (J mol ⁻¹ K ⁻¹)	$\Delta S^{\ddagger E}$ (J mol ⁻¹ K ⁻¹)
0.0000	27.2913	0.0000	54.0510	0.0000
0.0506	23.3913	-2.9489	42.5847	-8.9873
0.0997	21.2705	-4.1471	36.6759	-12.5255
0.1515	19.6866	-4.7577	32.4468	-14.2536
0.2000	18.6722	-4.8608	29.9622	-14.3966
0.2497	17.5465	-5.0526	27.1020	-14.8572
0.2909	16.7239	-5.1011	25.0898	-14.8803
0.3510	15.4886	-5.2071	22.0106	-15.0577
0.3999	14.5425	-5.2343	19.6820	-15.0254
0.4494	13.2905	-5.5563	16.3532	-15.9643
0.5003	12.2634	-5.6270	13.7346	-16.1254
0.5491	11.5964	-5.3771	12.1639	-15.3400
0.6002	11.2537	-4.7595	11.5958	-13.4410
0.6500	10.9117	-4.1658	10.9493	-11.6830
0.7000	10.9162	-3.2218	11.3642	-8.8541
0.7499	11.0027	-2.1977	11.9936	-5.8155
0.7999	10.8957	-1.3651	12.0344	-3.3606
0.8500	10.4188	-0.9007	10.8960	-2.0801
0.9000	9.4002	-0.9798	8.0123	-2.5498
0.9500	8.4272	-1.0133	5.1984	-2.9496
1.0000	8.5010	0.0000	5.7340	0.0000

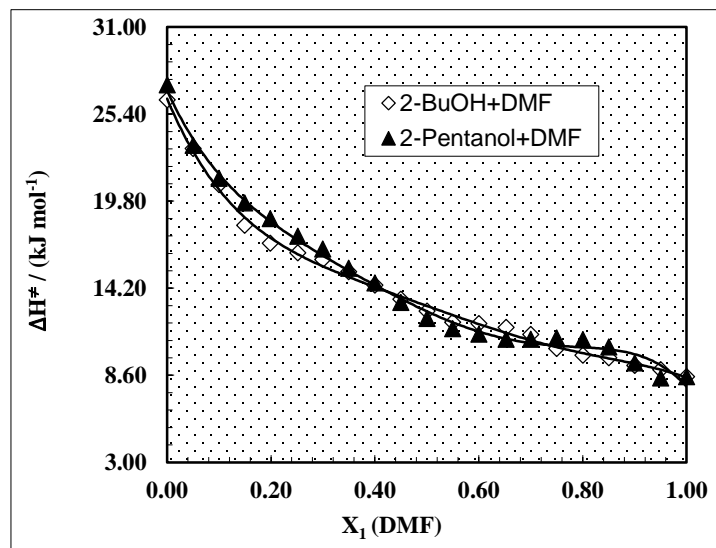


Figure 2: Comparative study of enthalpy of activation (ΔH^\ddagger) of 2-BuOH(x_2) + DMF(x_1) and 2-PnOH(x_2) + DMF(x_1) systems for different molar ratios at 298.15 K and 323.15 K.

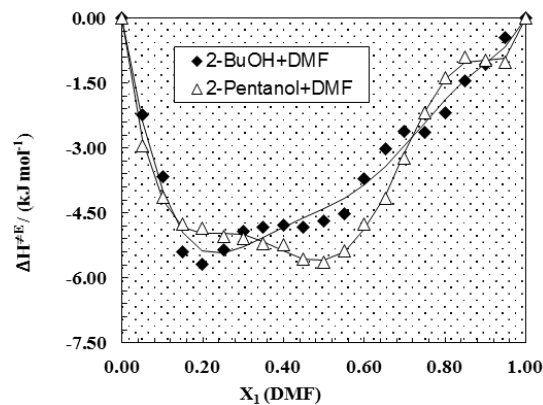


Figure 3: Comparative study of excess enthalpy of activation ($\Delta H^{\ddagger E}$) of 2-BuOH(X_2) + DMF(X_1) and 2-PnOH (X_2) + DMF(X_1) systems for different molar ratios at 298.15K and 323.15K.

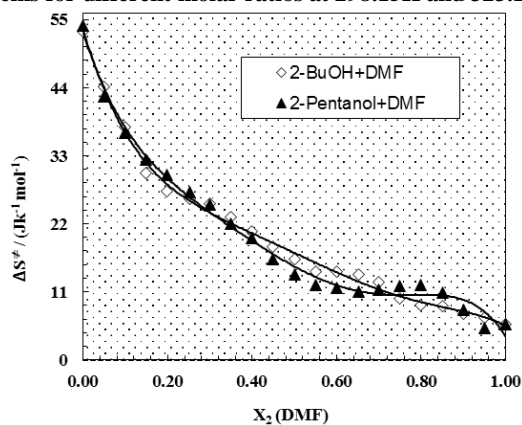


Figure 4: Comparative study of entropy of activation (ΔS^{\ddagger}) of 2-BuOH(X_2) + DMF(X_1) and 2-PnOH(X_2) + DMF(X_1) systems for different molar ratios at 298.15 K and 323.15 K.

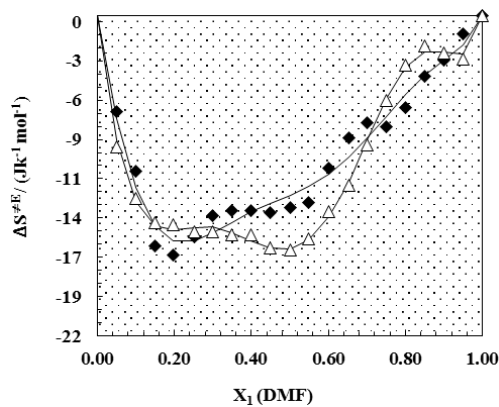


Figure 5: Comparative study of excess entropies of activation ($\Delta S^{\ddagger E}$) 2-BuOH(X_2) + DMF(X_1) and 2-PnOH(X_2) + DMF(X_1) systems for different molar ratios at 298.15 K and 323.15 K.

Interaction Parameter

According to Fort and Moore⁵⁴ Grunberg-Nission interaction parameter, ϵ may also be termed as an approximate index for estimating the strength of interaction between dissimilar components of a mixture during its flow. The Grunberg-Nissan interaction parameter (ϵ) was calculated by following the equation, where all the terms have their usual significances.

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 \epsilon \quad (9)$$

The values of ϵ are listed along with η and $\Delta\eta$ for the respective system as in Table 4 & 5 and Figure 6.

Table 4: Interaction parameter, ϵ , of the systems DMF(x_1) + 2-BuOH(x_2) for different molar ratios at temperatures 298.15 K - 323.15 K.
DMF+2-BuOH

X_1	298.15K	303.15K	308.15K	313.15K	318.15K	323.15K
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0506	-2.3317	-2.0290	-1.7426	-1.3484	-0.9350	-1.0106
0.0997	-2.1536	-1.9605	-1.6137	-1.2613	-0.9729	-1.0351
0.1515	-2.1946	-1.9769	-1.5265	-1.2849	-0.9887	-0.9816
0.2000	-2.0328	-1.8132	-1.4578	-1.1900	-0.9776	-1.0332
0.2497	-1.7922	-1.6235	-1.3436	-1.1532	-0.9698	-0.9823
0.2909	-1.6464	-1.4588	-1.2808	-1.1124	-0.9518	-0.9604
0.3510	-1.4767	-1.3741	-1.2134	-1.0642	-0.9258	-0.8454
0.3999	-1.3365	-1.2823	-1.0995	-1.0059	-0.8306	-0.7531
0.4494	-1.2625	-1.1996	-1.0347	-0.9584	-0.7584	-0.6916
0.5003	-1.1763	-1.1009	-0.9862	-0.8773	-0.7223	-0.7457
0.5491	-1.0795	-0.9980	-0.8879	-0.8106	-0.6449	-0.5074
0.6002	-0.9681	-0.8795	-0.7847	-0.6887	-0.5883	-0.4859
0.6500	-0.8999	-0.7865	-0.7016	-0.6293	-0.5415	-0.4817
0.7000	-0.8281	-0.7313	-0.6718	-0.5717	-0.4869	-0.4502
0.7499	-0.7841	-0.7183	-0.5798	-0.5173	-0.3608	-0.3981
0.7999	-0.6895	-0.1532	-0.1375	-0.0438	0.0178	0.1491
0.8500	-0.6069	-0.6352	-0.5622	-0.4370	-0.2937	-0.3362
0.9000	-0.6204	-0.6455	-0.4947	-0.4744	-0.2015	-0.3779
0.9500	-0.4102	-0.7027	-0.4540	-0.3021	-0.2533	-0.3115
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 5: Interaction parameter, ϵ , of the systems DMF(x_1) + 2-PnOH(x_2) for different molar ratios at temperatures 298.15 K - 323.15 K.

DMF+2-PnOH

X_1	298.15K	303.15K	308.15K	313.15K	318.15K	323.15K
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0506	-2.2302	-1.9268	-1.4206	-1.0502	-0.5430	-0.4474
0.0997	-1.8757	-1.5888	-1.2714	-1.0032	-0.6128	-0.4908
0.1515	-1.6424	-1.3743	-1.1535	-0.9036	-0.6137	-0.5278
0.2000	-1.5054	-1.2362	-1.0752	-0.8706	-0.6527	-0.5643
0.2497	-1.4146	-1.1614	-1.0291	-0.8416	-0.6608	-0.5708
0.2909	-1.3868	-1.1299	-1.0088	-0.8334	-0.6861	-0.6054
0.3510	-1.3448	-1.1159	-1.0203	-0.8460	-0.7045	-0.6244
0.3999	-1.3178	-1.1409	-1.0366	-0.8709	-0.7338	-0.6374
0.4494	-1.3297	-1.1800	-1.0776	-0.8828	-0.7382	-0.6489
0.5003	-1.3455	-1.2142	-1.0942	-0.9098	-0.7597	-0.7457
0.5491	-1.3229	-1.2074	-1.0920	-0.9341	-0.7705	-0.6643
0.6002	-1.2792	-1.1755	-1.0756	-0.9263	-0.7620	-0.6876
0.6500	-1.2562	-1.1225	-1.0066	-0.8971	-0.7399	-0.7068
0.7000	-1.1613	-1.0425	-0.9706	-0.8719	-0.7135	-0.7058
0.7499	-1.0322	-0.9732	-0.8685	-0.8494	-0.6817	-0.6937
0.7999	-0.9341	-0.8846	-0.8666	-0.8845	-0.7034	-0.6545
0.8500	-0.9188	-0.8888	-0.8580	-0.8216	-0.7540	-0.6849
0.9000	-1.0169	-0.9661	-0.9314	-0.8217	-0.7317	-0.6821
0.9500	-1.1904	-1.0604	-1.0042	-0.7256	-0.6791	-0.4941
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

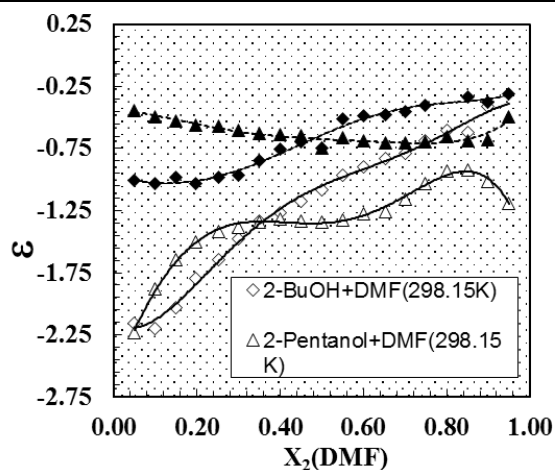


Figure 6: Comparative study of Interaction parameter (ϵ) of 2-BuOH(x_2) + DMF(x_1) and 2-PnOH(x_2) + DMF(x_1) systems for different molar ratios at 298.15 K and 323.15 K.

The observations regarding ΔH^\ddagger & ΔS^\ddagger and from their excess values found the follows results:

The values of ΔH^\ddagger & ΔS^\ddagger for both the systems are positive in the whole range of composition and decreases with the addition of DMF in alcohols (2-BuOH and 2-PnOH). The

order of increment of these viscous parameters follows: 2-PnOH > 2-BuOH > DMF for the pure components and 2-PnOH + DMF > 2-BuOH + DMF for their mixtures. Both $\Delta H^{#E}$ and $\Delta S^{#E}$ values show negative values with minima nearly at $x_2 \sim 0.20$ for 2-BuOH + DMF system and $x_2 \sim 0.50$ for 2-PnOH + DMF. The depth of minima is almost similar in type.

Liquids having high η values are expected to be associated with high $\Delta H^\#$, $\Delta S^\#$. Thus, associated liquids are generally characterized by large $\Delta H^\#$, $\Delta S^\#$ indicating that each of the activated species of this type need high energies in order to overcome high energy barriers. On the other hand, $\Delta H^\#$, $\Delta S^\#$ values for all the non-associated or non-polar liquids obviously should have very low values. This is just in accordance to the observed increasing order of $\Delta H^\#$, $\Delta S^\#$: 2-PnOH > 2-BuOH > DMF and also extends to the systems in the whole region of concentration as: 2-PnOH + DMF > 2-BuOH + DMF. As the values of interaction parameter, ϵ , are all negative at all compositions it leads to suggest that, intermolecular interaction of any type between amide DMF with 2-BuOH and 2-PnOH turns to be weaker or non-specific during flow. Also, negative values of $\Delta H^{#E}$, $\Delta S^{#E}$ their concentration dependences thus indicate that, all the 'activated' species within the mixtures of 2-PnOH + DMF and 2-BuOH + DMF require less energy (negative $\Delta H^{#E}$) but greater order (negative $\Delta S^{#E}$) compared to the 'inactivated' species, which were also in full support of negative interaction parameter, ϵ .

CONCLUSIONS

In conclusion, the molecular interaction of binary mixtures of *N,N*-dimethylformamide with 2-butanol and 2-pentanol have been measured at atmospheric pressure and at temperatures $T = 298.15-323.15$ K over the whole composition range. The viscosity and some of the thermodynamic parameters & their excess properties of binary mixtures of *N,N*-dimethylformamide with 2-butanol and 2-pentanol have been discussed. The Redlich-Kister polynomial equation was used to correlate the results. From the experimental data, it was found that the negative values due to rupture of H-bonding in alcohol-DMF (weak association) during flow, which was also supported by positive $\Delta H^\#$, $\Delta S^\#$ or $\Delta G^\#$ and their negative excess properties.

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DISCLOSURE STATEMENT

No conflict of interest was reported by the authors to publish this paper.

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