

Some Thermodynamic Properties of binary Mixtures of Alcohol isomers and Sulfolane at 298.15K

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Abstract :

The refractive indices, n_D densities ρ , and viscosities η of binary mixtures of sulfolane + n-butanol + sec-butanol + iso-butanol + tert-butanol + n-propanol and iso-propanol were measured at 298.15K. From experimental data, excess molar volum V^E , excess molar refractivity Δn_D , excess molar viscosity η^E and excess molar Gibbs free energy of activation of viscous flow ΔG^{*E} were calculated. From n-propanol – sulfolane and iso-propanol sulfolane mixtures showed negative Δn_D , n-butanol – sulfolane, sec-butanol – sulfolane, iso-butanol – sulfolane and tert-butanol sulfolane, Δn_D was positive over the whole mole fraction rang, while V^E , η^E and ΔG^{*E} show a negative deviation. The results obtained for binary mixtures suggest two types of molecular interaction. One is the formation of new stable complexes between the sulfone group of sulfolane and the hydroxyl group of alcohols and the second is related to the participation in destroying the mixture structure and forming a new structure. Excess molar quantities of these binary mixtures were found to be affected directly by the position of hydroxyl group and the steric associated with the methyl group. Key Word : Viscosity , density ,refractive index , sulfolane, excess properties and Mono alcohols.

Introduction

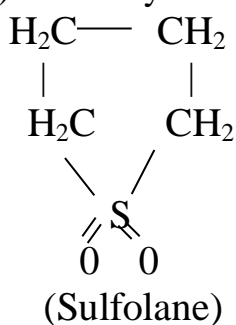
Studies of physicochemical properties of binary mixtures of considerable importance in the fundamental understanding of the nature of the interactions between unlike molecules. In recent years there has been considerable interest in theoretical and experimental investigations of the excess thermodynamic properties of binary mixtures [4-6]. Refractive index and density measurement are expected to shed some light on both solvent – solvent and solute – solvent interaction [7].

The excess properties of binary liquid mixtures are important for understanding and interpreting the interactions between molecules of a mixture [8].

The petroleum industry has given much attention to sulfolane , this interest has resulted from high selectivity and solvency for low molar mass monocyclic aromatic hydrocabons namely benzene , toluene , xylenes. Sulfolane as an effective extraction is widely accepted in practice for the recovery of aromatic hydrocarbons from petroleum products such as reformed naphthas and hydrogenated pyrolysis gasoline. The technological importance of sulfolane lead many researchers to study the binary and ternary mixtures containing sulfolane as a common solvent [9].

Sulfolane (tetrahydrothiophene-1,1-dioxide or tetra methylene sulfolane) is a highly dense , highly polar aprtic solvent [1-3]. It has been used in petroleum industry for the recovery of high purity moncyclic aromatic hydrocarbons particulary benzene , toluene and xylene from naphthas , hydrogenated pyrolysis gasolines.

Sulfolane is a globular molecule and with a firly high dipole moment (4.8D) with only the negative and the dipole exposed.



Experimental

Liquid Materials:

The following liquid materials were used are :

Name of Material	Purity	Name of Company
Sulfolane (tetra methylene sulfone)	+ 99.5 %	Fluka
1 – butanol (n – butanol)	+ 99 %	Aldrich
2 – butanol (sec-butanol)	+ 99 %	Aldrich
2- methyl-1-propanol (iso-butanol)	+ 99 %	Aldrich
2- methyl-2-propanol (tert-butanol)	+ 99 %	Aldrich
1-propanol(n-propanol)	+ 99.5 %	Aldrich
2-propanol (iso-propanol)	+ 99.5 %	Aldrich

Table (1) : Densities ρ , Refractive indices n_D and viscosities η of alcohols and sulfolane.

Liquid	ρ (gm . cm ⁻³)		n_D		η (cP)	
	Obs.	Lit.	Obs.	Lit.	Obs.	Lit.
n-butanol	0.8054	0.8060	1.3981	1.3977	2.641	2.635
sec- butanol	0.8025	0.8026	1.3964	1.3960	3.518	3.522
iso- butanol	0.7977	0.7977	1.3938	1.3939	3.865	3.910
Tert- butanol	0.7810	0.7812	1.3855	1.3851	3.989	3.975
n-propanol	0.7993	0.7999	1.3842	1.3839	2.121	1.980
iso-propanol	0.7801	0.7808	1.3760	1.3755	2.224	2.125
sulfolane	1.2651	1.2655	1.4832	1.4838	11.605	11.599

Obs. = observed

Lit.=Literature

Density measurements:

Densities of pure components and binary mixtures were measured with an Anton paar digital densimeter (model DMA 60/602) with an accuracy of $\pm 10^{-5}$ gcm⁻³ . Air and double calibration of the densimeter. Experiments were generally repeated at least three times for each composition and the results were averaged.

Viscosity measurements:

The kinematic Viscosity ($\nu = \eta / \rho$) of pure solvents and their mixture were measured by using (cannan – ubbelohde semi micro) viscometer , the uncertainty in the kinematic viscosity was ± 0.001 mpa.s. All measurement was performed in a thermostat maintained at desired temperature with a accuracy of ± 0.001 k. It is described above and performed at least three times , and the results were averaged to give the final values.

Refractive index measurement

The refractive indices of pure liquids and their binary mixture were measured using a thermostatted Abbe refractometer. We calibrated the refractometer by measuring the refractive indices of doubly distilled water , various temperatures. The values of refractive indices were obtained using sodium D light. The temperature of the test liquids between the prisms of the refractometre during the measurement was maintained to an uncertainty of ± 0.001 K by circulating water through the jacket around the prisms from an electronically controlled thermostatic water bath and the temperature was measured with a digital. The uncertainty in refractive index measurement was within ± 0.0001 The temperature of the test liquids during the measurement was maintained to an uncertainty of ± 0.01 K in a electrically controlled thermostatic water bath.

Results and discussion :

The experimental densities ρ viscosities η , refractive indices n_D , excess molar Volumes V^E , Excess molar viscosities η^E and excess molar refractivity Δn_D of binary mixtures of alcohols isomers with sulfolane at 298. 15K are listed in tables [2].

The corresponding excess properties were calculated as follows

Excess molar volumes V^E :

The experimental excess molar volume, V^E result obtained from density measurements of the binary mixtures of sulfolane + alcohols isomers were determined from the following equation

$$V^E = (x_1 m_1 + x_2 m_2) / \rho_m - x_1 m_1 / \rho_1 - x_2 m_2 / \rho_2 \dots\dots\dots(1)$$

Where m_1 , x_1 and ρ_1 are respectively the molecular weight , mole fraction and density of the pure sulfolane , m_2 , x_2 and ρ_2 represent the , molecular weight , mole fraction and density of alcohol , (ρ_m) is the density of the binary mixtures , the obtained excess molar volum of binary mixtures of sulfolane + alcohols at 298.15k were plotted as function of mole fraction (x_1) of sulfolane figure (2). All V^E are negative for sulfolane + alcohols at 298.15K whole mole fraction rang.

Excess molar viscosities η^E

Excess molar viscosities η^E were obtained form viscosity measurement of the binary mixtures of sulfolane + alcohols isomers and where determined from the following equation

$$\eta^E = \eta_m - (x_1 \eta_1 + x_2 \eta_2) \dots\dots\dots(2)$$

Where x_1 , η_1 are respectively the mole fraction and viscosity of sulfolane , x_2 and η_2 represent the mole fraction and viscosity of alcohol. (η_m) is the viscosity of the binary mixture. The obtained excess molar viscosities of binary mixtures of sulfolane + alcohols at 298.15K were plotted as function of mole fraction (x_1) of sulfolane figure (3) . It is observed in all binary mixtures , that η^E is negative.

Excess molar activation energies , ΔG^{*E}

Excess molar activation energies, ΔG^{*E} have been calculated by using the experimental results of viscosity (η) and density of the pure component liquids binary mixtures by following equation.

$$\Delta G^{*E} = RT [\ln \eta_m V_m - (X_1 \ln \eta_1 V_1 + X_2 \ln \eta_2 V_2)] \dots\dots (3)$$

Where η_m and V_m are respective the viscosity and molar volume of the binary mixture , x_1 , η_1 and V_1 represent the mole fraction , viscosity and molar volum of the pure sulfolane x_2 , η_2 and V_2 are respectively the mole fraction, viscosity and molar volum of alcohol.

The obtained excess molar activation energies of binary mixtures of sulfolane + alcohols at 298.15k. were plotted as function of mole fraction (x_1) of sulfolane figure (4). ΔG^{*E} is negative over the whole mole fraction.

Excess molar refractivity Δn_D .

Lorentz – Loernz excess molar refractivity , Δn_D has been calculated using the experimental results. of refractive index (n_D) of the pure liquids and the binary mixtures by the following equation [10].

$$\Delta n_D = n_D - \phi_1 n_{D1} - \phi_2 n_{D2} \dots \dots \dots (4)$$

Where n_D is the molar rfractivities of binary mixtures , and n_{D1} , n_{D2} represent the molar refrativities of component liquids 1 and 2.

The ϕ_i is volume fraction of ith component liguids , the volum fraction $\sum_{i=1}^2 \phi_i = 1$

Was calculated by using

$$\phi_i = X_i V_i / \sum_{i=1}^2 X_i V_i \dots \dots \dots (5)$$

Where X_i and V_i is the mole fraction and molar volume of the pare component liquids in the mixture from Lorentz - Lorenz quation n_{D1} , n_{D2} and n_D were calculated :

$$n_{D1} = \left[\frac{n_1^2 - 1}{n_1^2 + 2} \right] \left[\frac{m_1}{p_1} \right] \dots \dots \dots (6)$$

$$n_{D2} = \left[\frac{n_2^2 - 1}{n_2^2 + 2} \right] \left[\frac{m_2}{p_2} \right] \dots \dots \dots (7)$$

where ρ_1 , n_1 and m_1 are respectively the density , refractive index and molecular weight of the pure sulfolane , and ρ_2 n_2 and m_2 are respectively the density, refractive index and molecular weight of the pure alcohol.

$$n_D = \left[\frac{n_m^2 - 1}{n_m^2 + 2} \right] \cdot V_m \dots \dots \dots (8)$$

Where n_m is the refractive index of the binary mixures. V_m is the molar volume which where calculated by using

$$V_m = \frac{X_1 m_1 + X_2 m_2}{p_m} \dots \dots \dots (9)$$

Where p_m is the density of the binary mixture.

The calculated values of excess molar refractivity, Δn_D of binary mixturs of sulfolane + alcohols at , 298.15K were presented graphically in figures (1). It is obseved in binary mixtures containg n-propanol and iso-propaol, Δn_D is negative and changed to positive in all binay mixtures containing n-butanol , sec – butand , iso butanol and tert – butanol.

Discussion

It was established that alcohols in Liquid exists in self- associated dynamic – linear chain structure or cyclic dimers , established by hydrogen

bonding, the extent of such association in various alcohols is determined by the position of hydroxyl group (OH) , the carbon chain length and branching of the alkyl chain of which can enhance the steric hindrance in the monomeric molecules.

For n- propanol – sulfolane and iso- propanol- sulfolane mixtures, Δn_D is negative this behavior suggests that the molecular interaction between the hydroxyl group of n-propanol and sulfone group of sulfolane is greater than the molecular interaction between n-propanol molecules , this leads to destroying the self associated structure of n- propanol by globular sulfolane and giving the highest negative values of Δn_D . In case of iso-propanol – sulfolane mixtures , it seems that the position of hydroxyl group deactivates the molecular interaction , therefore Δn_D is less negative than n-propanol – sulfolane mixtures. As the number of carbon atoms increase in the chain of alcohol and the position of hydroxyl group change , Δn_D becomes positive , for all the butyl alcohols isomers mixtures, excess molar volume V^E for all binary mixtures studied here were negative over the whole mole fraction range. In case of n- propanol – sulfolane mixtures , V^E is the highest negative such behavior may be explained qualitatively by postulating that a strong hydrogen bond complexes formed through the sulfone group of sulfolane and the hydroxyl group of n- propanol. Comparing the V^E result of iso- propanol , which has the same number of carbon atoms but the position of hydroxyl group (OH) is different. It is observed in this mixture , V^E is less negative . This is probably due to the effect of crowding of methyl group (steric effect). As the chain of hydrocarbon increase and also the position of hydroxyl group V^E is less negative and this appears sharply in case of mixtures of tert – butanol – sulfolane which has the lowest negative V^E due to the high (steric effect) of methyl groups , Excess molar viscosity η^E and excess activation energy of viscous flow ΔG^{*E} are negative over the whole mole by new fraction range at 298.15K. Such behavior obtained that can be explained complexes are formed through strong hydrogen bonding between the hydroxyl group of alcohol and sulfone group S destroying the self-association of alcohols and geometrical and steric factors in alcohols , this hypothesis is substantiated by the negative value of mixing obtained. It is apparent that the magnitude of η^E and ΔG^{*E} for n-propanol is smaller than other alcohols, therefore the degree to which such hydrogen bond complexes are lessened

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as a result of introducing sulfolane molecules. As a result we observed η^E and ΔG^{*E} for tert- butanol mixture are the highest negative values.

Table (2) :Experimental Values Of density , viscosity and refractive index and Calculated Values of excess Molar Volume , excess Viscosity , excess Gibbs free energy and excess refractive index for binary Mixtures.

X_1 Sulfolane + X_2 n – butanol at 298.15 K

X_1	ρ (gm.cm ⁻³)	V^E (Cm ⁻³ .Mol ⁻¹)	η (CP)	η^E (CP)	$\Delta G^* \times 10^3$ (J.Mol ⁻¹)	n_D	Δn_D (Cm ⁻³ .Mol ⁻¹)
0.0000	0.8054	0.0000	2.641	0.000	0.00	1.3981	0.0000
0.0977	0.8508	- 0.0808	2.877	-0.041	- 0.10	1.4086	0.0687
0.1917	0.8954	- 0.1300	3.245	- 0.078	- 0.20	1.4174	0.0940
0.2922	0.9429	- 0.1555	3.708	- 0.093	- 0.23	1.4265	0.1064
0.3859	0.9868	- 0.1637	4.221	- 0.102	- 0.26	1.4349	0.1204
0.4883	1.0345	- 0.1711	4.873	- 0.110	- 0.28	1.4438	0.1310
0.5893	1.0811	- 0.1623	5.698	- 0.103	- 0.26	1.4524	0.1268
0.7009	1.1345	- 0.1468	6.806	- 0.091	- 0.23	1.4615	0.1146
0.7937	1.1745	- 0.1389	7.901	- 0.071	- 0.20	1.4689	0.0976
0.9159	1.2296	- 0.0761	9.672	- 0.035	- 0.09	1.4784	0.0724
1.0000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000

X_1 Sulfolane + X_2 Sec – butanol at 298.15 K

X_1	ρ (gm.cm ⁻³)	V^E (Cm ³ .Mol ⁻¹)	η (CP)	η^E (CP)	$\Delta G^* \times 10^3$ (J.Mol ⁻¹)	n_D	Δn_D (C m ³ .Mol ⁻¹)
0.0000	0.8025	0.0000	3.518	0.000	0.00	1.3964	0.0000
0.0975	0.8488	- 0.0503	3.688	- 0.039	- 0.11	1.4069	0.0664
0.1924	0.8940	- 0.0988	4.044	- 0.090	- 0.19	1.4159	0.0893
0.2875	0.9389	- 0.1336	4.473	- 0.103	- 0.26	1.4244	0.0940
0.3800	0.9824	- 0.1431	4.929	- 0.116	- 0.29	1.4329	0.1103
0.4790	1.0287	- 0.1502	5.509	- 0.123	-0.31	1.4419	0.1207
0.5855	1.0782	- 0.1534	6.320	- 0.113	- 0.28	1.4508	0.1122
0.6857	1.1244	- 0.1350	7.221	- 0.099	- 0.25	1.4594	0.1062
0.7904	1.1724	- 0.1012	8.289	- 0.079	- 0.22	1.4682	0.0987
0.8902	1.2178	- 0.0498	9.525	- 0.045	- 0.12	1.4762	0.0778
1.0000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000
0.0000	0.7977	0.0000	3.865	0.000	0.00	1.3938	0.0000
0.0986	0.8440	- 0.0197	3.939	- 0.048	- 0.12	1.4052	0.0298
0.1933	0.8893	- 0.0417	4.278	- 0.085	- 0.22	1.4142	0.0569
0.2886	0.9348	- 0.0705	4.680	- 0.124	- 0.29	1.4230	0.0701
0.3832	0.9796	- 0.0853	5.152	- 0.134	- 0.33	1.4325	0.0876
0.4938	1.0318	- 0.0887	5.760	- 0.144	- 0.36	1.4417	0.0991
0.5831	1.0738	- 0.0986	6.406	- 0.136	- 0.34	1.4494	0.0912
0.6894	1.1233	- 0.0841	7.273	- 0.126	- 0.30	1.4581	0.0754
0.7908	1.1702	- 0.0615	8.236	- 0.093	- 0.24	1.4668	0.0665
0.9004	1.2206	- 0.0327	9.458	- 0.054	- 0.12	1.4756	0.0399
1.0000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000

X_1 Sulfolane + X_2 iso – butanol at 298.15 K

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X₁ Sulfolane + X₂ tert – butanol at 298.15 K

X ₁	ρ (gm.cm ⁻³)	V ^E (CM ³ .Mol ⁻¹)	η (CP)	η ^E (CP)	ΔG* x10 ⁻³ (J.Mol ⁻¹)	n _D	Δ n _D (Cm ⁻³ .Mol ⁻¹)
0.0000	0.7810	0.0000	3.989	0.000	0.00	1.3855	0.0000
0.0989	0.8301	- 0.0191	4.021	- 0.051	- 0.13	1.3960	0.0269
0.1988	0.8778	- 0.0389	4.382	- 0.104	- 0.23	1.4054	0.0410
0.2991	0.9266	- 0.0516	4.811	- 0.136	- 0.32	1.4155	0.0617
0.4012	0.9763	- 0.0656	5.316	- 0.145	- 0.35	1.4257	0.0740
0.4961	1.0225	- 0.0778	5.837	- 0.152	- 0.37	1.4352	0.0831
0.6144	1.0798	- 0.0661	6.689	- 0.141	- 0.35	1.4466	0.0753
0.7016	1.1220	- 0.0551	7.391	- 0.134	- 0.33	1.4580	0.0671
0.8006	1.1698	- 0.0346	8.310	- 0.095	- 0.24	1.4690	0.0554
0.8838	1.1200	- 0.0210	9.188	- 0.065	- 0.13	1.4760	0.0382
1.0000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000

X₁-Sulfolane + X₂ iso – propanol at 298.15 K

X ₁	ρ (gm.cm ³)	V ^E (M ³ .Mol ⁻¹)	η (CP)	η ^E (CP)	ΔG* x10 ⁻³ (J.Mol ⁻¹)	n _D	Δ n _D (M ³ .Mol ⁻¹)
0.0000	0.7993	0.0000	2.121	0.000	0.00	1.3842	0.0000
0.0969	0.8487	- 0.0716	2.433	- 0.027	- 0.07	1.3946	- 0.1332
0.1789	0.8945	- 0.1101	2.731	- 0.051	- 0.12	1.4066	- 0.2170
0.2820	0.9494	- 0.1524	3.213	- 0.064	- 0.15	1.4201	- 0.2840
0.3731	0.9959	- 0.1738	3.722	- 0.072	- 0.17	1.4308	- 0.3164
0.4722	1.0442	- 0.1844	4.356	- 0.085	- 0.18	1.4412	- 0.3209
0.5797	1.0942	- 0.1926	5.275	- 0.074	- 0.17	1.4515	- 0.3032
0.6884	1.1421	- 0.1750	6.404	- 0.065	- 0.15	1.4601	- 0.2565
0.7890	1.1844	- 0.1200	7.755	- 0.045	- 0.10	1.4670	- 0.1825
0.8579	1.2124	- 0.0902	8.828	- 0.032	- 0.08	1.4713	- 0.1349
1.000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000

X ₁	ρ (gm.cm ³)	V ^E (CM ³ .Mol ⁻¹)	η (CP)	η ^E (CP)	ΔG* x10 ⁻³ (J.Mol ⁻¹)	n _D	Δ n _D (Cm ⁻³ .Mol ⁻¹)
0.0000	0.7801	0.0000	2.224	0.000	0.00	1.3760	0.0000
0.0956	0.8363	- 0.0675	2.461	- 0.057	- 0.13	1.3760	- 0.0987
0.1826	0.8855	- 0.1014	2.775	- 0.080	- 0.19	1.4010	- 0.1734
0.2752	0.9359	- 0.1514	3.186	- 0.095	- 0.23	1.4140	- 0.2379
0.3653	0.9830	- 0.170	3.643	- 0.110	- 0.26	1.4251	- 0.2587
0.4744	1.0375	- 0.1766	4.326	- 0.118	- 0.27	1.4375	- 0.2605
0.5843	1.0898	- 0.1704	5.234	- 0.109	- 0.26	1.4486	- 0.2550
0.6504	1.1200	- 0.1555	5.891	- 0.100	- 0.24	1.4546	- 0.2300
0.7705	1.1730	- 0.1211	7.297	- 0.085	- 0.20	1.4645	- 0.1710
0.8756	1.2173	- 0.0752	8.870	- 0.063	- 0.14	1.4719	- 0.1121
1.0000	1.2651	0.0000	11.605	0.000	0.00	1.4832	0.0000

X₁-Sulfolane + X₂ n – propanol

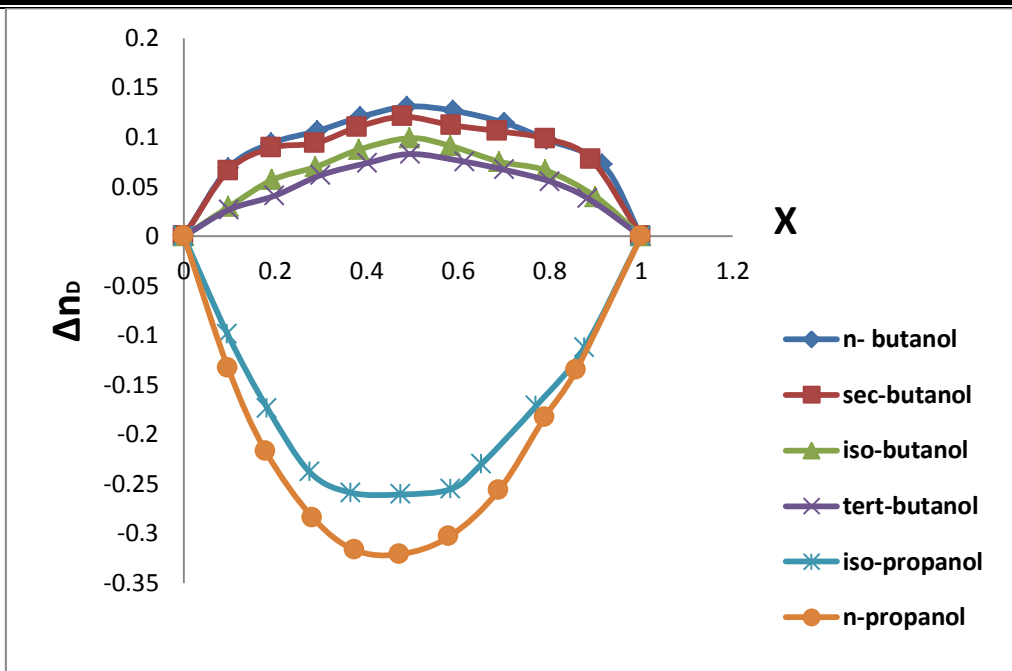


Fig 1: Excess molar refractivity Δn_D for X sulfolane +(1-X) isomers alcohols at 298.15K.

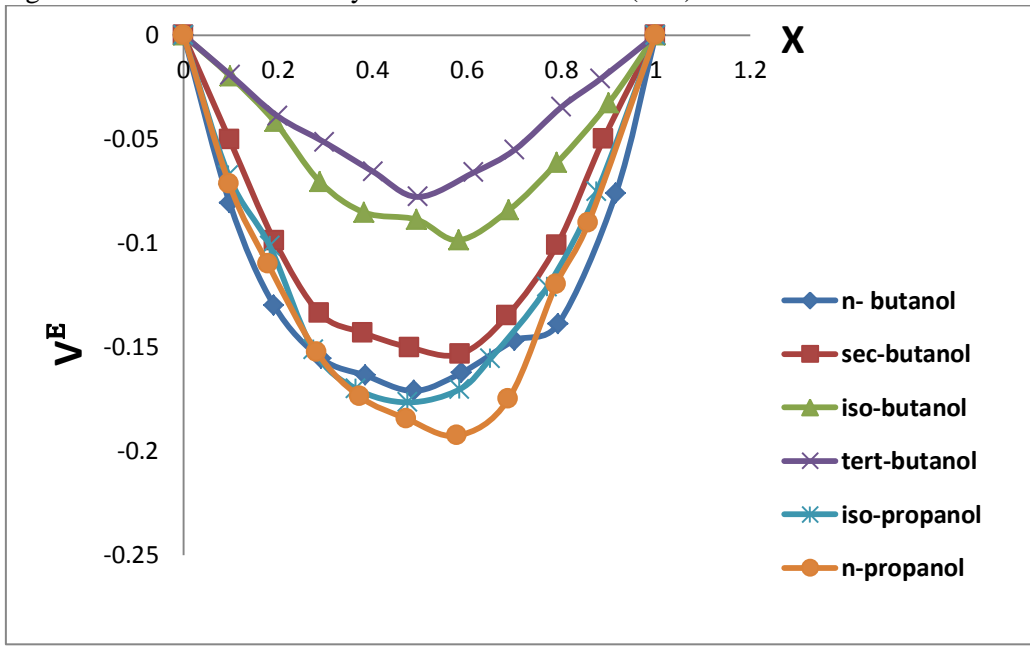


Fig 2: Excess molar volume V^E for X sulfolane +(1-X) isomers alcohols at 298.15K.

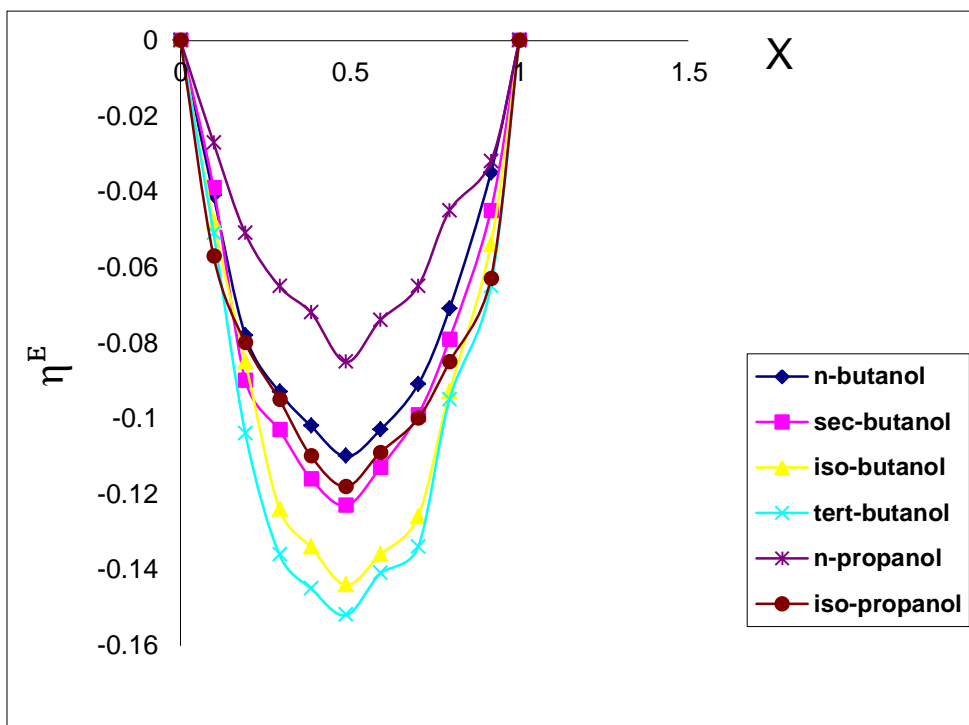


Fig 3: Excess molar viscosity η^E for X sulfolane +(1-X) isomers alcohols at 298.15K.

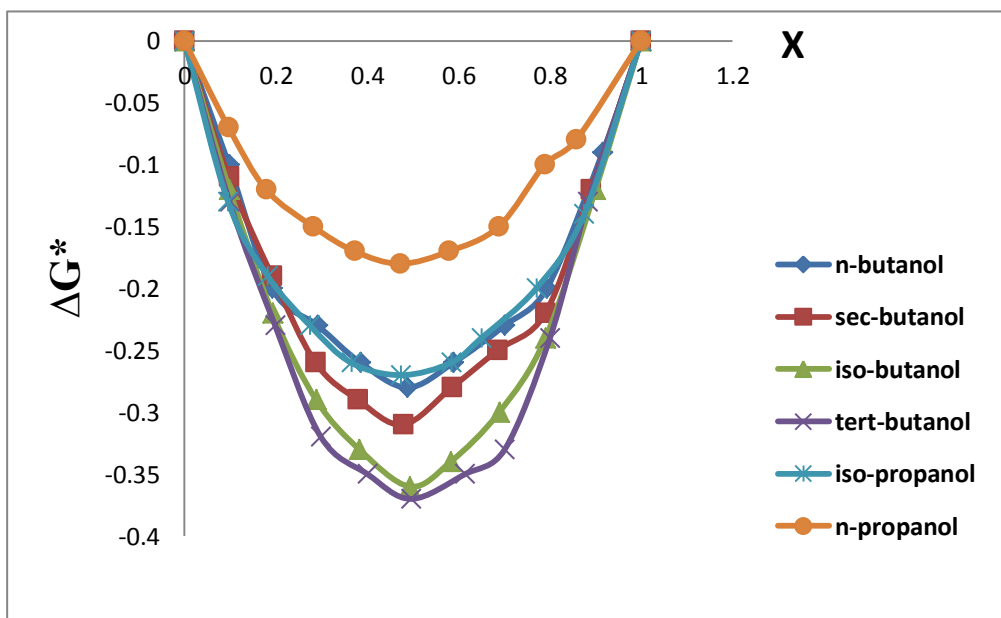


Fig4: Excess molar Gibbs free energy of activation of viscous flow ΔG^* for X sulfolane +(1-X) isomers alcohols at 298.15K.

Conclusions

We conclude that this excess function is effected directly by the position of hydroxyl group and steric effects associated with methyl groups.

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بعض الخصائص الترموديناميكية لمخاليط ثنائية المكون من ايزومرات الكحولات والسلفولان في درجة 298.15 كلفن

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الخلاصة :

ان الاهمية الصناعية للسلفولان كمذيب قطبي تكمن في كونه ذو كفاءة عالية في انتقاء الهيدروكربونات العطرية احادية الحلقة (البنزين ، التلويين والزايلينات) من المشتقات النفطية حيث انه يستخدم على نطاق واسع في استخلاص الهيدروكربونات العطرية من الريفورميت والكازولين والنافثا الخفيفة ، تم في الدراسة دراسة الخواص الترموديناميكية لبعض المخاليط الثنائية لهذا المذيب حيث تم قياس الكثافة ، معامل الانكسار واللزوجة علمياً للمخاليط ثنائية المكون من السلفولان والكحولات، - بيوتانول ، - بيوتانول ، - ميثيل - - بروبانول ، - ميثيل - - بروبانول عند درجة حرارة 298.15 كلفن.

تم حساب الحجم المولارية الفائقة V^E من القياسات العلمية للكثافة وحساب الانكسارية المولارية الفائضة Δn_D من القياسات العملية لمعامل الانكسار بينما حسبت اللزوجة المولارية الفائضة η^E وطاقة التنشيط الفائضة ΔG^{*E} من القياسات العملية لكل من اللزوجة والكثافة وذلك على المدى الكلي للكسر المولي ، ان الفائدة الاساسية من هذه الدراسة هي معرفة التأثيرات الجزئية بين السلفولان والكحولات وبيان تأثير الاشكال الجزئية على هذه التأثيرات وانواعها.

اتضح من نتائج دراسة الخواص الفائضة لهذه المخاليط الثنائية ان هناك نوعين من التداخلات الجزئية ، الاول يتكون نتيجة تكوين معقدات جديدة من خلال تكوين اصرة هيدروجينية قوية بين مجموعة السلفون في مذيب السلفولان ومجموعة الهيدوكسل في الكحول والثاني يتكون نتيجة تكسير التركيب الاساس للمادة وتكوين تركيب جديد كذلك فان الدوال الفائضة تتأثر مباشرة بموقع مجموعة الهيدروكسيل في الكحول والاعاقة الفراغية من قبل مجموعة الميثيل.

الكلمات المفتاحية : اللزوجة ، الكثافة ، معامل الانكسار ، السلفولان ، الدوال الترموديناميكية الفائضة والكحولات