Measurement and modeling of transport properties of binary liquid mixtures containing oxygenates and n-alkanes

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Abstract−Dynamic viscosities (η) of the binary liquid mixtures of 2-propanol (1)+n-alkanes (C_6, C_7, C_9) (2) at T=288.15 K to 303.15 K and ethanol (1)+n-alkanes (C_6, C_7, C_8) (2) at T=308.15 K to 318.15 K were experimentally measured over the whole composition range. Experimental values of η were used to compute the deviation in dynamic viscosity ($\Delta\eta$) and these $\Delta\eta$ values were correlated with the Redlich-Kister equation. The η values of binary mixtures were also calculated using several empirical correlations and mixing rules like Grunberg-Nissan, Tamura-Kurata, Kati-Chaudhari and McLaughlin-Ubbelohde and found that the Grunberg-Nissan correlation gave the best estimation. The $\Delta\eta$ values were also predicted by an approach given by Singh et al. [Indian J Chem 29, 263 (1990)].

Keywords: Dynamic Viscosity, Deviation in Dynamic Viscosity, Ethanol, 2-Propanol, Alkanes, Correlations

INTRODUCTION

The thermodynamic properties of a binary mixture of alkanol and hydrocarbon are very helpful in understanding the intermolecular interaction, which is very useful for petrochemical industries and for formulating motor fuel [1-5]. Among all the thermodynamic properties of a liquid mixture, viscosity is one of the most significant property [6-8] as viscosity of pure and mixture fluids is needed for designing different processes like hydraulic calculations, mass transfer, fluid flow and heat transfer [9-13] in various chemical industries. This paper is a continuation of our previous work [14]. In this research paper, we investigated experimental data of dynamic viscosity (η) of binary liquid systems containing 2-propanol and n-alkanes (C_6, C_7, C_9) from 288.15 K to 303.15 K. Dynamic viscosity (η) of binary liquid systems containing ethanol and n-alkanes (C_6, C_7, C_8) at 308.15 K and 318.15 K were also measured over the entire composition range. These results were further used to calculate the deviation in dynamic viscosity ($\Delta\eta$). The molecular collaboration of alkanols with alkanes has its own great importance due to various industrial applications [15]. As 2-propanol and ethanol are rich oxygenates, highly soluble, self-associating components therefore useful in petrochemical industries and used as a gasoline stabilizer to increase the efficiency of gasoline engines. The data obtained by this research work is valuable for designing and developing environmentally friendly fuel, petrochemical additives and formulating diesel and gasoline [16,17]. The experimental values of $\Delta\eta$ for binary mixture of 2-propanol with n-hexane at 298.15 K were found to agree well with literature data [18], while the data of $\Delta\eta$ for the other binary systems were not found in literature at the studied temperature.

EXPERIMENTAL SECTION

2-Propanol and n-nonane were supplied by Sigma Aldrich,

Table 1. Chemicals specifications

S. No.	Sample	CAS No.	Make	Mass fraction purity	Purification method
1.	Ethanol	64-17-5	Merck	≥0.999	Distillation
2.	2-Propanol	67-63-0	Sigma-Aldrich	≥0.995	Distillation
3.	n-Hexane	110-54-3	Merck	≥0.985	Distillation
4.	n-Heptane	142-82-5	Merck	≥0.990	Distillation
5.	n-Octane	111-65-9	Merck	≥0.990	Distillation
6.	n-Nonane	111-84-2	Sigma-Aldrich	≥0.990	Distillation

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Table 2. Purities, measured densities (ρ), viscosities (η) and refractive indices (n_D) of the pure components

Compound	T/K	$\rho/10^3 \times$g cm^{-3}		η/mPa s		n	
		This work	Literature	This work	Literature	This work	Literature
2- Propanol	288.15	792.20	789.22[19]	2.763	------	1.3794	------
	293.15	785.07	785.28[20]	2.382	------	1.3773	------
	298.15	780.74	780.56[21]	2.046	2.061[18]	1.3752	1.3753[16]
	303.15	776.61	776.76[22]	1.767	1.724[23]	1.3730	------
n-Hexane	288.15	664.40	663.91[24]	0.358	------	------	------
	293.15	659.91	659.57[25]	0.341	------	1.3761	1.3756[26]
	298.15	655.38	654.9[27]	0.322	0.313[18]	1.3734	1.37234[28]
			655.10[28]				
	303.15	650.82	650.37[29]	0.305	0.309[30]	1.3708	------
					0.285[31]		
n-Heptane	288.15	688.08	688.20[24]	0.453	------	1.3903	------
	293.15	683.85	683.92[25]	0.432	------	1.3878	1.3878[26]
	298.15	679.60	679.40[28]	0.409	0.400[32]	1.3852	1.38512[28]
	303.15	675.33	675.47[25]	0.389	0.387[30]	1.3826	------
					0.368[31]		
n-Octane	288.15	706.78	------	0.583	------	1.3918	------
	293.15	702.76	702.62[25]	0.554	0.545[33]	1.3928	------
	298.15	698.71	698.60[18]	0.522	0.522[32]	1.3951	1.39519[34]
					0.518[18]		
	303.15	694.66	694.50[29]	0.496	0.478[31]	1.3925	------
n-Nonane	288.15	721.73	------	0.748	0.768[35]	------	------
	293.15	717.87	717.85[25]	0.699	0.716[35]	1.4056	1.4058[26]
	298.15	713.97	714.01[25]	0.652	0.669[35]	1.4032	1.40326[34]
	303.15	710.07	710.11[25]	0.612	0.620[31]	1.4009	------
Ethanol	298.15	787.08	785.90[36]	1.087	1.0991[36]	1.3612	----
	308.15	778.41	777.94[33]	0.957	0.910[33]	1.3566	1.3553[33]
	318.15	769.57	769.15[33]	0.802	-----	1.3520	1.3512 [33]

while other chemicals were supplied by Merck. and were simply distilled and then kept over molecular sieves (4 Å) for more than 72 hours. Their specifications are given in Table 1. To ensure the purity of compounds, density (ρ) and refractive indices (n) of pure compounds were measured using Anton Paar DSA-5000M and Abbemat-200 with an uncertainty of $\pm10^{-6}$ g cm^{-3} and $\pm1\times10^{-4}$, respectively. The η of pure and binary mixtures was measured with LOVIS 2000 ME microviscometer integrated with Anton Paar

DSA-5000M with accuracy ±0.005 mPa·s and temperature was controlled by built-in Peltier thermostat with temperature repeatability of 0.005 K. All sample mixtures were prepared using an electronic balance (OHAUS, AR224CN) having ±0.1 mg accuracy. The estimated averaged standard uncertainty (u(η)) was calculated in the manner described elsewhere [7] and found to be less than 0.001 mPa·s. The experimental values of ρ, n and η for pure components are mentioned in Table 2 and these agree well with the literature values [18-36], while for mixtures, η values are reported in Table 3.

Table 3. Experimental viscosities (η/mPa s), deviation in viscosities ($\Delta\eta_{exptl}$/mPa s), calculated viscosities ($\Delta\eta^{\dagger}$) and ($\Delta\eta^{*}$) from Eq. (4) and (5) of binary liquid mixtures

x_1	η	$\Delta\eta_{exptl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
2-Propanol (1)+n-hexane (2)				
T=288.15 K				
0.0000	0.358	0.0000	0.0000	0.0000
0.1125	0.370	−0.2587	−0.1788	−0.5264
0.1504	0.384	−0.3358	−0.3547	−0.6653
0.2838	0.428	−0.6126	−0.7688	−0.9523
0.4115	0.511	−0.8367	−0.9133	−0.9791

Table 3. Continued

x_1	η	$\Delta\eta_{exptl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
0.5237	0.660	−0.9574	−0.9482	−0.9121
0.6463	0.876	−1.0365	−0.9997	−0.8519
0.7067	1.046	−1.0117	−1.0416	−0.8385
0.8010	1.313	−0.9710	−1.0917	−0.8026
0.8685	1.631	−0.8162	−1.0557	−0.7061
0.9373	2.130	−0.4821	−0.8669	−0.4581
1.0000	2.763	0.0000	0.0000	0.0000

Table 3. Continued

x_1	η	$\Delta\eta_{exp\,tl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
		T=293.15 K		
0.0000	0.341	0.0000	0.0000	0.0000
0.1125	0.355	−0.2157	−0.0553	−0.5485
0.1504	0.368	−0.2800	−0.2150	−0.6552
0.2838	0.402	−0.5183	−0.5776	−0.8367
0.4115	0.479	−0.7019	−0.7486	−0.8395
0.5237	0.587	−0.8228	−0.8198	−0.7654
0.6463	0.803	−0.8572	−0.8354	−0.6239
0.7067	0.924	−0.8595	−0.8213	−0.5330
0.8010	1.161	−0.8148	−0.7746	−0.3674
0.8685	1.446	−0.6676	−0.7294	−0.2372
0.9373	1.844	−0.4100	−0.6842	−0.1056
1.0000	2.382	0.0000	0.0000	0.0000
		T=298.15 K		
0.0000	0.322	0.0000	0.0000	0.0000
0.1125	0.329	−0.1870	−0.0347	−0.4602
0.1504	0.347	−0.2343	−0.1717	−0.5478
0.2838	0.380	−0.4313	−0.4835	−0.6950
0.4115	0.447	−0.5845	−0.6362	−0.6984
0.5237	0.518	−0.7068	−0.7038	−0.6385
0.6463	0.681	−0.7553	−0.7193	−0.5178
0.7067	0.793	−0.7474	−0.7054	−0.4381
0.8010	1.021	−0.6819	−0.6607	−0.2919
0.8685	1.252	−0.5677	−0.6206	−0.1795
0.9373	1.681	−0.2569	−0.5880	−0.0729
1.0000	2.046	0.0000	0.0000	0.0000
		T=303.15 K		
0.0000	0.305	0.0000	0.0000	0.0000
0.1125	0.309	−0.1606	−0.0332	−0.3816
0.1504	0.331	−0.1941	−0.1431	−0.4543
0.2838	0.370	−0.3500	−0.3932	−0.5774
0.4115	0.418	−0.4886	−0.5165	−0.5823
0.5237	0.497	−0.5736	−0.5723	−0.5355
0.6463	0.624	−0.6260	−0.5874	−0.4396
0.7067	0.744	−0.5940	−0.5773	−0.3754
0.8010	0.914	−0.5619	−0.5417	−0.2557
0.8685	1.107	−0.4677	−0.5074	−0.1615
0.9373	1.373	−0.3026	−0.4757	−0.0687
1.0000	1.767	0.0000	0.0000	0.0000
		2-Propanol (1)+n-heptane (2)		
		T=288.15 K		
0.0000	0.453	0.0000	0.0000	0.0000
0.0945	0.471	−0.2003	−0.0106	−0.3999
0.1389	0.485	−0.2888	−0.2155	−0.5580
0.2234	0.496	−0.4731	−0.5285	−0.7835
0.3286	0.542	−0.6700	−0.7726	−0.9217
0.4217	0.614	−0.8132	−0.8826	−0.9399
0.5105	0.695	−0.9372	−0.9354	−0.9065
0.6557	0.936	−1.0316	−0.9994	−0.8297
0.7515	1.195	−0.9938	−1.0394	−0.7769
0.8313	1.473	−0.9003	−1.0317	−0.6926

Table 3. Continued

x_1	η	$\Delta\eta_{exp\,tl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
0.9501	2.238	−0.4096	−0.7745	−0.3265
1.0000	2.763	0.0000	0.0000	0.0000
		T=293.15 K		
0.0000	0.432	0.0000	0.0000	0.0000
0.0945	0.441	−0.1753	0.0493	−0.4320
0.1389	0.461	−0.2418	−0.1340	−0.5603
0.2234	0.479	−0.3886	−0.3926	−0.7153
0.3286	0.509	−0.5637	−0.6024	−0.7977
0.4217	0.567	−0.6874	−0.7208	−0.8041
0.5105	0.638	−0.7894	−0.7891	−0.7663
0.6557	0.840	−0.8706	−0.8142	−0.6189
0.7515	1.041	−0.8563	−0.7741	−0.4659
0.8313	1.301	−0.7520	−0.7145	−0.3125
0.9501	1.957	−0.3276	−0.6207	−0.0791
1.0000	2.382	0.0000	0.0000	0.0000
		T=298.15 K		
0.0000	0.409	0.0000	0.0000	0.0000
0.0945	0.429	−0.1347	0.0415	−0.3601
0.1389	0.430	−0.2063	−0.1093	−0.4649
0.2234	0.441	−0.3337	−0.3209	−0.5900
0.3286	0.475	−0.4719	−0.4942	−0.6569
0.4217	0.539	−0.5604	−0.5948	−0.6640
0.5105	0.590	−0.6546	−0.6543	−0.6350
0.6557	0.761	−0.7213	−0.6753	−0.5121
0.7515	0.931	−0.7081	−0.6373	−0.3799
0.8313	1.152	−0.6178	−0.5830	−0.2474
0.9501	1.698	−0.2663	−0.5075	−0.0555
1.0000	2.046	0.0000	0.0000	0.0000
		T=303.15 K		
0.0000	0.389	0.0000	0.0000	0.0000
0.0945	0.401	−0.1182	0.0215	−0.2967
0.1389	0.407	−0.1734	−0.1016	−0.3839
0.2234	0.411	−0.2858	−0.2748	−0.4890
0.3286	0.447	−0.3948	−0.4164	−0.5466
0.4217	0.497	−0.4732	−0.4981	−0.5545
0.5105	0.546	−0.5464	−0.5462	−0.5325
0.6557	0.688	−0.6045	−0.5626	−0.4348
0.7515	0.845	−0.5795	−0.5305	−0.3279
0.8313	1.023	−0.5115	−0.4834	−0.2187
0.9501	1.482	−0.2162	−0.4106	−0.0534
1.0000	1.767	0.0000	0.0000	0.0000
		2-Propanol (1)+n-nonane (2)		
		T=288.15 K		
0.0000	0.748	0.0000	0.0000	0.0000
0.1056	0.766	−0.1948	−0.0892	−0.3693
0.2190	0.773	−0.4163	−0.4197	−0.6154
0.3463	0.831	−0.6148	−0.6544	−0.7568
0.4659	0.899	−0.7878	−0.7915	−0.8072
0.5683	1.007	−0.8860	−0.8681	−0.8100
0.6561	1.142	−0.9280	−0.9060	−0.7849
0.7344	1.308	−0.9199	−0.9087	−0.7318

Table 3. Continued

x_1	η	$\Delta\eta_{exp\,tl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
0.8067	1.511	−0.8625	−0.8691	−0.6411
0.9136	2.008	−0.5810	−0.6852	−0.3830
0.9658	2.316	−0.3781	−0.5131	−0.1756
1.0000	2.763	0.0000	0.0000	0.0000
		T=293.15 K		
0.0000	0.699	0.0000	0.0000	0.0000
0.1056	0.731	−0.1457	0.0190	−0.3576
0.2190	0.718	−0.3496	−0.3198	−0.5879
0.3463	0.766	−0.5158	−0.5558	−0.7023
0.4659	0.807	−0.6761	−0.6804	−0.7126
0.5683	0.915	−0.7404	−0.7363	−0.6707
0.6561	0.961	−0.8422	−0.7559	−0.6064
0.7344	1.099	−0.8360	−0.7517	−0.5275
0.8067	1.331	−0.7257	−0.7257	−0.4324
0.9136	1.750	−0.4867	−0.6304	−0.2351
0.9658	2.003	−0.3214	−0.5484	−0.1034
1.0000	2.382	0.0000	0.0000	0.0000
0.0000	0.699	0.0000	0.0000	0.0000
		T=298.15 K		
0.0000	0.652	0.0000	0.0000	0.0000
0.1056	0.652	−0.1472	−0.0552	−0.3144
0.2190	0.668	−0.2893	−0.3223	−0.4967
0.3463	0.711	−0.4237	−0.4955	−0.5794
0.4659	0.716	−0.5855	−0.5825	−0.5840
0.5683	0.830	−0.6141	−0.6167	−0.5491
0.6561	0.922	−0.6446	−0.6202	−0.4944
0.7344	1.039	−0.6368	−0.6022	−0.4251
0.8067	1.222	−0.5546	−0.5652	−0.3416
0.9136	1.541	−0.3846	−0.4674	−0.1765
0.9658	1.738	−0.2603	−0.3979	−0.0750
1.0000	2.046	0.0000	0.0000	0.0000
		T=303.15 K		
0.0000	0.612	0.0000	0.0000	0.0000
0.1056	0.614	−0.1200	−0.0238	−0.2730
0.2190	0.623	−0.2420	−0.2283	−0.4066
0.3463	0.665	−0.3470	−0.3608	−0.4592
0.4659	0.715	−0.4351	−0.4420	−0.4650
0.5683	0.755	−0.5133	−0.4887	−0.4474
0.6561	0.834	−0.5358	−0.5089	−0.4121
0.7344	0.932	−0.5283	−0.5061	−0.3599
0.8067	1.089	−0.4548	−0.4825	−0.2905
0.9136	1.358	−0.3093	−0.4071	−0.1473
0.9658	1.512	−0.2155	−0.3541	−0.0611
1.0000	1.767	0.0000	0.0000	0.0000
		Ethanol (1)+n-hexane (2)		
		T=308.15 K		
0.0000	0.2970	0.0000	0.0000	0.0000
0.0423	0.3038	−0.0212	0.0230	−0.0958
0.0731	0.3131	−0.0321	0.0012	−0.1451
0.1349	0.3222	−0.0638	−0.0288	−0.2061
0.2545	0.3394	−0.1256	−0.0568	−0.2416

Table 3. Continued

x_1	η	$\Delta\eta_{exp\,tl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
0.4113	0.3581	−0.2104	−0.0742	−0.2350
0.5212	0.4083	−0.2327	−0.0836	−0.2227
0.5884	0.4547	−0.2306	−0.0873	−0.2094
0.7429	0.5889	−0.1984	−0.0835	−0.1453
0.7735	0.6114	−0.1961	−0.0805	−0.1263
0.8149	0.6596	−0.1752	−0.0755	−0.0983
0.9620	0.8818	−0.0501	−0.0611	−0.0075
1.0000	0.9570	0.0000	0.0000	0.0000
		T=318.15 K		
0.0000	0.2820	0.0000	0.0000	0.0000
0.0423	0.2884	−0.0155	0.0133	−0.0826
0.0731	0.2938	−0.0262	−0.0132	−0.1246
0.1349	0.3010	−0.0511	−0.0484	−0.1753
0.2545	0.3195	−0.0949	−0.0773	−0.2010
0.4113	0.3277	−0.1682	−0.0914	−0.1903
0.5212	0.3710	−0.1821	−0.0988	−0.1782
0.5884	0.3988	−0.1891	−0.1010	−0.1666
0.7429	0.5036	−0.1647	−0.0916	−0.1129
0.7735	0.5288	−0.1554	−0.0870	−0.0972
0.8149	0.5685	−0.1372	−0.0798	−0.0742
0.9620	0.7300	−0.0522	−0.0601	−0.0032
1.0000	0.8020	0.0000	0.0000	0.0000
		Ethanol (1)+n-heptane (2)		
		T=308.15 K		
0.0000	0.3690	0.0000	0.0000	0.0000
0.0511	0.3626	−0.0364	−0.0093	−0.0739
0.1461	0.3856	−0.0693	−0.1032	−0.1543
0.2322	0.3841	−0.1214	−0.1479	−0.1866
0.3469	0.4117	−0.1612	−0.1780	−0.2002
0.4291	0.4306	−0.1907	−0.1907	−0.2011
0.5270	0.4760	−0.2029	−0.2017	−0.1980
0.6178	0.5242	−0.2081	−0.2075	−0.1908
0.7247	0.6009	−0.1942	−0.2034	−0.1713
0.8191	0.6928	−0.1578	−0.1826	−0.1368
0.9086	0.7836	−0.1196	−0.1412	−0.0825
0.9563	0.8776	−0.0538	−0.1086	−0.0430
1.0000	0.9570	0.0000	0.0000	0.0000
		T=318.15 K		
0.0000	0.3380	0.0000	0.0000	0.0000
0.0511	0.3390	−0.0227	0.0088	−0.0596
0.1461	0.3389	−0.0669	−0.0718	−0.1231
0.2322	0.3521	−0.0937	−0.1107	−0.1476
0.3469	0.3750	−0.1239	−0.1379	−0.1565
0.4291	0.3934	−0.1437	−0.1502	−0.1559
0.5270	0.4190	−0.1636	−0.1616	−0.1521
0.6178	0.4600	−0.1647	−0.1685	−0.1450
0.7247	0.5166	−0.1577	−0.1678	−0.1280
0.8191	0.5802	−0.1379	−0.1538	−0.1003
0.9086	0.6508	−0.1088	−0.1252	−0.0590
0.9563	0.7228	−0.0589	−0.1030	−0.0303
1.0000	0.8020	0.0000	0.0000	0.0000

Table 3. Continued

x_1	η	$\Delta\eta_{exp\,tl}$	$\Delta\eta^{\dagger}$	$\Delta\eta^{*}$
		Ethanol (1)+n-octane (2)		
		T=308.15 K		
0.0000	0.4680	0.0000	0.0000	0.0000
0.0568	0.4662	−0.0296	−0.0184	−0.0582
0.1097	0.4774	−0.0443	−0.0593	−0.0946
0.1951	0.4709	−0.0925	−0.1012	−0.1290
0.3033	0.4925	−0.1239	−0.1301	−0.1485
0.3855	0.5186	−0.1380	−0.1440	−0.1550
0.4825	0.5474	−0.1566	−0.1564	−0.1588
0.5882	0.5884	−0.1673	−0.1652	−0.1582
0.6833	0.6398	−0.1623	−0.1652	−0.1497
0.8047	0.7230	−0.1386	−0.1457	−0.1192
0.9025	0.8239	−0.0854	−0.1072	−0.0718
0.9655	0.8919	−0.0482	−0.0696	−0.0283
1.0000	0.9570	0.0000	0.0000	0.0000
		T=318.15 K		
0.0000	0.4230	0.0000	0.0000	0.0000
0.0568	0.4187	−0.0258	−0.0179	−0.0530
0.1097	0.4308	−0.0337	−0.0538	−0.0857
0.1951	0.4228	−0.0741	−0.0897	−0.1157
0.3033	0.4337	−0.1043	−0.1131	−0.1313
0.3855	0.4512	−0.1180	−0.1236	−0.1356
0.4825	0.4730	−0.1329	−0.1326	−0.1372
0.5882	0.5060	−0.1400	−0.1385	−0.1349
0.6833	0.5456	−0.1364	−0.1373	−0.1262
0.8047	0.6152	−0.1128	−0.1199	−0.0990
0.9025	0.6994	−0.0656	−0.0880	−0.0586
0.9655	0.7767	−0.0122	−0.0578	−0.0228
1.0000	0.8020	0.0000	0.0000	0.0000

RESULTS AND DISCUSSION

The $\Delta\eta$ values calculated using Eq. (1) are reported in Table 3 as well as shown in Fig. 1 and Fig. 2.

$$\Delta\eta_{exp.} = \eta_m - \sum_{j=1}^{n} x_j \eta_j \qquad (1)$$

where η_j, η_m represent experimental dynamic viscosities of pure component j and binary mixture, respectively. 'n' denotes number of components whereas x_j is mole fraction of j component in binary mixture. The experimental values for $\Delta\eta$ were correlated with following Redlich-Kister Eq. [37]:

$$\Delta\eta_{cal.} = x_1(1-x_1)\left[\sum_{n-1}^{4} X^{(n)}(2x_1-1)^{(n-1)}\right] \qquad (2)$$

where $X^{(n)}$ denotes adjustable parameters and mentioned in Table 4 together with standard deviations determined by Eq. (3).

$$\sigma(\Delta\eta) = \left\{\left[\sum\left(\Delta\eta_{exp.(Eq.\,(1))} - \Delta\eta_{calcd\,(Eq.\,(2))}\right)^2\right]/(x-y)\right\}^{1/2} \qquad (3)$$

where y symbolizes the number of adjustable parameters and x is the number of experimental data points. The experimental values $\Delta\eta$ for 2-propanol (1)+hexane (2) at 298.15 K were found to com-
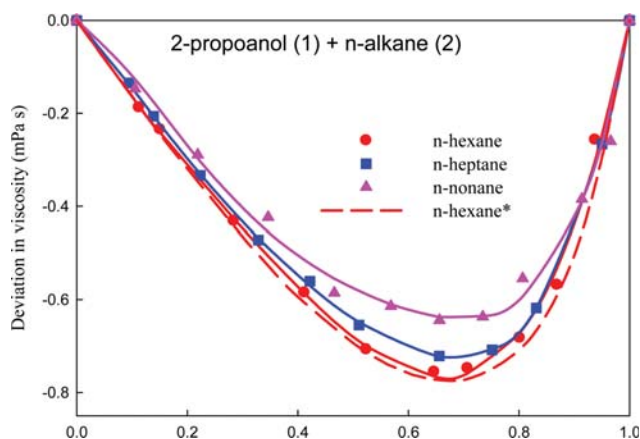

Fig. 1. Deviation in viscosity of binary mixtures of 2-propanol (1)+ n-alkanes (2) as functions of mole fraction of 2-propanol at 298.15 K; symbols represent experimental values and lines represent values calculated from Redlich-Kister equation, dashed lines represent the literature values *Ref. [18]. as functions of mole fraction of 2-propanol at 298.15 K.
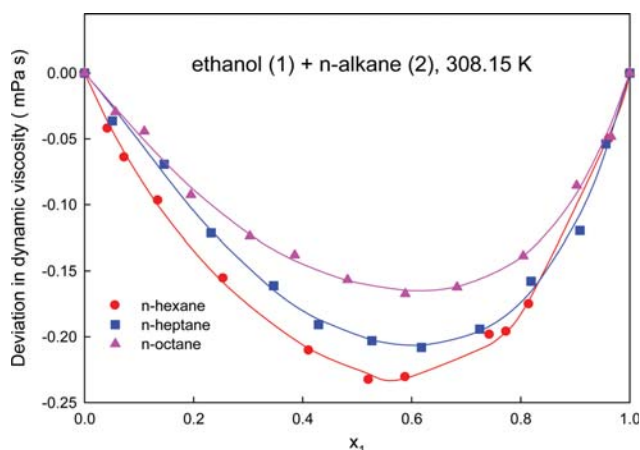

Fig. 2. Deviation in viscosity of binary mixtures of ethanol (1)+n-alkanes (2) as functions of mole fraction of 2-propanol at 308.15 K; symbols represent experimental values and lines represent values calculated from Redlich-Kister equation.

pare well with literature data [18] as shown in Fig. 1. While the $\Delta\eta$ data for other binary systems were not found in literature at studied temperature.

The $\Delta\eta$ values were found to be negative for all studied binary systems over the whole composition range. An organized pattern of $\Delta\eta$ with increase in chain length of n-alkanes may be seen in Fig. 1. The magnitude of negative values of $\Delta\eta$ for 2 propanol (1)+ n-alkane (2) follows the order: n-nonane<n-heptane<n-hexane. The negative values of $\Delta\eta$ indicate the presence of weak interactions between unlike molecules. It is also due to rupture of self-associated bonds in alkanols. The magnitude of negative values is decreasing with increase of chain length of alkane, which may be due to weakening of the interaction between unlike molecules. It is due to (i) rupture of hydrogen bonding of 2-propanol having self-association characteristics and a non-associating forces present in n-alkane and (ii) the existence of dispersal forces among two mixing

Table 4. Values of adjustable parameters of Redlich-Kister Equation and standard deviation ($\sigma(\Delta\eta)$) for viscosities at different temperatures

T/K	$X^{(0)}$	$X^{(1)}$	$X^{(2)}$	$X^{(3)}$	$\sigma(\Delta\eta)$
	2-Propanol (1)+n-hexane (2)				
288.15 K	−3.7113	−1.9386	−2.0203	−2.0271	0.0150
293.15 K	−3.1429	−1.6097	−1.5837	−1.6937	0.0124
298.15 K	−2.7168	−1.854	−1.0061	−0.0788	0.0254
303.15 K	−2.1948	−1.2282	−1.1371	−0.9948	0.0167
	2-Propanol (1)+n-heptane (2)				
288.15 K	−3.6469	−2.1425	−1.8834	−1.7241	0.0123
293.15 K	−3.0912	−1.9679	−1.4949	−1.0428	0.0068
298.15 K	−2.5606	−1.5942	−1.2284	−0.8707	0.0053
303.15 K	−2.1460	−1.2994	−0.9789	−0.6484	0.0046
	2-Propanol (1)+n-nonane (2)				
288.15 K	−3.2386	−1.7532	−2.0962	−2.5785	0.0417
293.15 K	−2.8037	−1.8432	−1.6535	−1.6870	0.0423
298.15 K	−2.3036	−1.1606	−1.1469	−1.5469	0.0398
303.15 K	−1.8461	−1.1071	−1.0857	−0.9050	0.0322
	Ethanol (1)+n-hexane (2)				
308.15 K	−0.9029	−0.2668	−0.2333	0.0755	0.0039
318.15 K	−0.7237	−0.1796	−0.1674	−0.1421	0.0047
	Ethanol (1)+n-heptane (2)				
308.15 K	−0.7970	−0.2663	−0.1999	−0.2638	0.0063
318.15 K	−0.6138	−0.2211	−0.3387	−0.3398	0.0048
	Ethanol (1)+n-octane (2)				
308.15 K	−0.6366	−0.1996	−0.2177	−0.1895	0.0046
318.15 K	−0.5485	−0.2109	−0.0451	0.0409	0.0054

compounds in binary systems. These observations support our findings in our earlier paper [14]. Viscosity deviation ($\Delta\eta$) values for ethanol (1)+alkanes (2) are presented in Fig. 2. The $\Delta\eta$ values at equimolar composition follow the order: n-octane<n-heptane<n-hexane. Negative $\Delta\eta$ value confirms specific interactions among unlike molecules. The $\Delta\eta$ value decreases with rise in temperature as shown in Fig. 3 and Fig. 4 due to weakening of intermolecular interactions with temperature rise.

The $\Delta\eta$ values were also predicted by using the following equation according to an approach given by Singh et al. [38]:

$$\Delta\eta = K^* V^E \tag{4}$$

The V^E data of these binary mixtures were already published in our previous research paper [14]. We used experimental data of $\Delta\eta$ and V^E for an equimolar composition ($x_1=0.5$) of binary mixtures to calculate K^* factors, which was consequently used to estimate the values of $\Delta\eta$ at other mole fraction [39-41].

Then a modified approach of the Singh equation was used. Alkanol is self-associated by hydrogen bond; thus the $\Delta\eta$ values of the current binary systems would include the following contributions due to (a) depolymerization of alkanol to give monomers. Initially, self-associated alkanol establishes (1_n)......(2) contacts with alkane. The change in viscosity would be directly proportional to the mole fraction of alkane (x_2). Therefore, $\Delta\eta_{(1_n)......(2)}=K'x_2$, where K' is a proportionality constant. Then (1_n)......(2) contacts cause depolymeriza-

Fig. 3. Deviation in viscosity of 2-propanol (1)+n-alkanes (2) as functions of mole fraction of 2-propanol at temperature (288.15, 293.15, 298.15, 303.15) K.

tion in alkanol (1_n) to yield monomers of alkanol, resulting in decrease in viscosity. This decrease in viscosity would be proportional to mole fraction of alkanol (x_1). Therefore, $-\Delta\eta_{depolym.}=K''x_1$, where K'' is a proportionality constant. If it was assumed that $K'=K''=K^\dagger$, then net change in viscosity due to depolymerization would be $\Delta\eta_{depolym.}=K^\dagger(x_2-x_1)$; (b) interaction of monomers of alkanol with alkane to form unlike interactions. After the process of depolymerization, both components act as a monomer, hence, the $\Delta\eta_{1\text{-}2\,int.}$ may be due to unlike (1-2) interaction [42]. The net $\Delta\eta$ of the current binary mixtures would be given by:

$$\Delta\eta = \Delta\eta_{depolym.} + \Delta\eta_{1\text{-}2\,int.}$$
$$= K^\dagger(x_2-x_1) + K^\sigma V^E \tag{5}$$
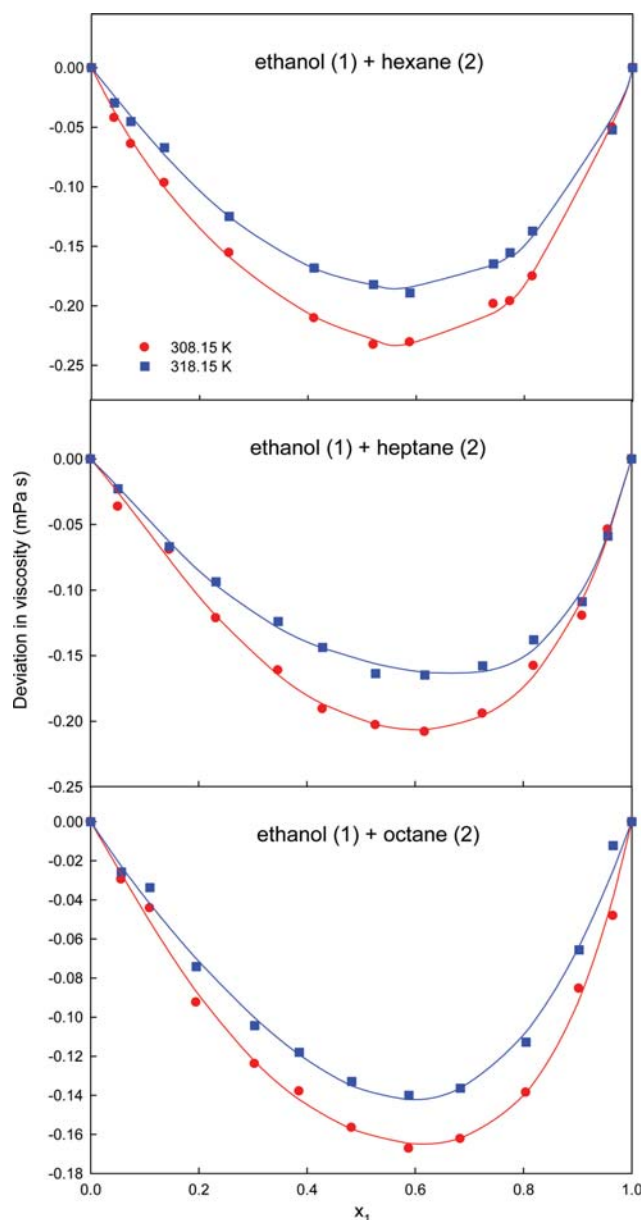
Rearranging above equation, we get

Fig. 4. Deviation in viscosity of ethanol (1)+n-alkanes (2) as functions of mole fraction of 2-propanol at temperature (308.15, 318.15) K.

$$\frac{\Delta\eta}{(x_2-x_1)}=K^o\frac{V^E}{(x_2-x_1)}+K^\dagger$$

The plot of $\Delta\eta/(x_2-x_1)$ versus $V^E/(x_2-x_1)$ must be linear, having intercept $K^\dagger$ and slope K^o as shown in Fig. 5 and Fig. 6, only if all the basic terms are valid in solving Eq. (5). The values of K^o, $K^\dagger$, r (correlation coefficient) for all binary systems are reported in Table 5. From Table 5 it is interesting to note that the value of the correlation coefficient, r, for all current binary mixtures is very near to one. This gives acceptance to all the assumptions for these mixtures $K'=K''=K^\dagger$. The values of K^o and $K^\dagger$ were used to estimate the viscosity change $\Delta\eta$ for binary mixtures. Such data are recorded in Table 3 and $\Delta\eta^\dagger$ values well agreed with their corresponding experimental $\Delta\eta$ values and their standard deviation $(\sigma^\dagger)$ is mentioned
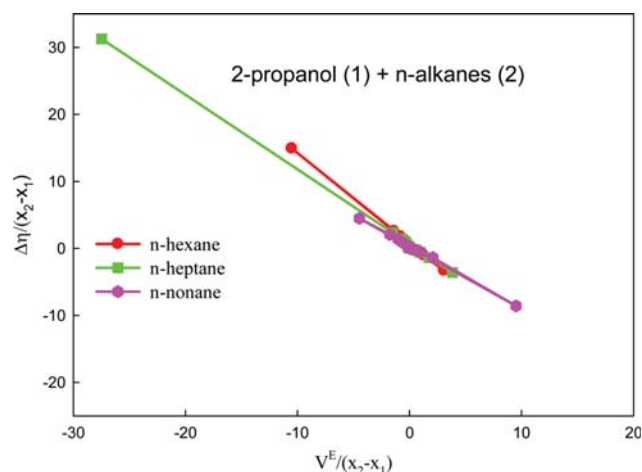
Fig. 5. Plots of $\Delta\eta/(x_2-x_1)$ versus $V^E/(x_2-x_1)$ for 2-propanol (1)+n-alkanes (2) at 298.15 K.
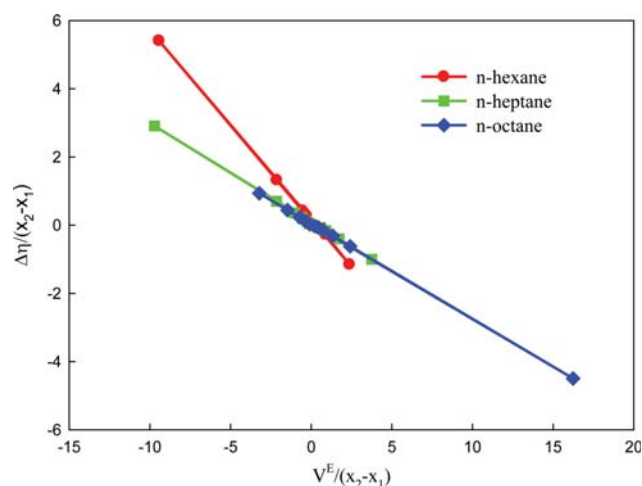

Fig. 6. Plots of $\Delta\eta/(x_2-x_1)$ versus $V^E/(x_2-x_1)$ for ethanol (1)+n-alkanes (2) at 308.15 K.

in Table 5. The K^o is known as intermolecular interactional factor, if K^o values are high then the degree of interaction is also high between unlike (1-2) molecules. Whereas, $K^\dagger$ values specify the degree of depolymerization because of unlike molecules interaction. The $\Delta\eta$ is the result of involvement of two factors that are $K^\dagger$ and K^o. It can be concluded that with growth in chain length of n-alkanes, the values of K^o are also growing (Table 5), which supports our results.

1. Correlations

The viscosity values of these binary systems were correlated by the following correlations.

Grunberg and Nissan (G-N) [43]

$$\eta=\exp\left[\sum_{i=1,2}(x_i\ln\eta_i)+G_{12}\prod_{i=1,2}x_i\right] \qquad (6)$$

Tamura and Kurata (T-K) [44]

$$\eta=\left[\sum_{i=1,2}x_i\phi_i\eta_i+2T_{12}\prod_{i=1,2}(x_i\phi_i)^{1/2}\right] \qquad (7)$$

Hind, McLaughlin and Ubbelohde [45]

Table 5. Interactional parameters (K*, K†, Kᵒ) of Eqs. (4) and (5), correlation coefficient r and standard deviations of viscosity (σ* and σ†) for binary mixture calculated from Eqs. (4) and (5) respectively

System	T/K	K*	K†	Kᵒ	r	σ*	σ†
2-Propanol (1)+n-hexane (2)	288.15	−1.6652	0.4592	−1.6914	0.9982	0.2057	0.1610
	293.15	−1.6583	0.6580	−1.7086	0.9991	0.3185	0.1087
	298.15	−1.2834	0.5841	−1.3591	0.9985	0.2736	0.1218
	303.15	−0.9748	0.4632	−1.0019	0.9991	0.2243	0.0743
2-Propanol (1)+n-heptane (2)	288.15	−1.4590	0.4902	−1.4891	0.9997	0.2066	0.1460
	293.15	−1.4325	0.6005	−1.4516	0.9998	0.2852	0.1272
	298.15	−1.1037	0.5013	−1.1190	0.9997	0.2370	0.1043
	303.15	−0.8423	0.3962	−0.8509	0.9998	0.1875	0.0829
2-Propanol (1)+n-nonane (2)	288.15	−1.1639	0.3602	−1.1767	0.9995	0.1678	0.0654
	293.15	−1.1765	0.4777	−1.1772	0.9981	0.2214	0.1079
	298.15	−0.9002	0.3436	−0.9340	0.9989	0.1702	0.0651
	303.15	−0.6687	0.3147	−0.6664	0.9982	0.1375	0.0641
Ethanol (1)+n-hexane(2)	308.15	−0.5562	0.0632	−0.2022	0.9848	0.0642	0.1048
	318.15	−0.3750	0.0632	−0.2022	0.9848	0.0603	0.0599
Ethanol (1)+n-heptane (2)	308.15	−0.3770	0.0719	−0.3766	0.9991	0.0396	0.0253
	318.15	−0.2507	0.0785	−0.2594	0.9989	0.0366	0.0194
Ethanol (1)+n-octane (2)	308.15	−0.2796	0.0445	−0.278	0.9998	0.0248	0.0117
	318.15	−0.2092	0.0382	−0.2043	0.9996	0.0247	0.0177

Table 6. Percentage standard deviation of viscosity of various correlations like Gruenberg-Nissan (G-N), Tamura-Kurata (T-K), Hind-McLaughlin-Ubbelohde (H-Mc-U) and Katti-Chaudhari (K-C) correlation and their international parameters

Binary mixture	T/K	G-N		T-K		H-Mc-U		K-C	
		σ (%)	G_{12}	σ (%)	T_{12}	σ (%)	H_{12}	σ (%)	W_{vis}/RT
2-Propanol (1)+n-hexane (2)	288.15	2.8521	−1.9547	26.3654	−0.2300	45.5951	−0.6037	7.9383	−4.4637
	293.15	2.6714	−1.8943	23.3717	−0.1393	40.4662	−0.4565	8.6583	−4.4038
	298.15	3.2201	−1.8823	20.9548	−0.0699	36.5073	−0.3398	10.5085	−4.3915
	303.15	3.8698	−1.7567	19.4502	−0.0103	33.0229	−0.2412	10.4928	−4.2655
2-Propanol (1)+n-heptane (2)	288.15	3.9377	−1.9426	18.8901	0.0104	35.710	−0.3757	13.1017	−4.3354
	293.15	4.6424	−1.8919	17.3812	0.0651	32.4595	−0.2622	13.9592	−4.2860
	298.15	4.6732	−1.7899	14.9891	0.1254	28.4122	−0.1530	13.9266	−4.1836
	303.15	4.0656	−1.7251	12.5599	0.1629	24.6338	−0.0747	13.3524	−4.1184
2-Propanol (1)+n-nonane (2)	288.15	6.9198	−1.8566	11.5560	0.3558	25.5516	−0.2059	10.3274	−28.7387
	293.15	8.7975	−1.8675	12.2967	0.3293	25.5383	−0.1434	20.4058	−28.8227
	298.15	6.0681	−1.7227	8.3506	0.3922	19.6872	−0.0068	19.9532	−27.8532
	303.15	6.3657	−1.5842	7.8859	0.4220	17.8349	−0.0845	19.4980	−26.9249
Ethanol (1)+n-hexane (2)	308.15	0.2720	−1.2276	3.9207	0.3155	5.5737	0.1416	6.5771	−3.6300
	318.15	0.0801	−1.1419	2.2112	0.2970	5.6241	0.1555	8.5079	−3.5430
Ethanol (1)+n-heptane (2)	308.15	0.0595	−1.0190	1.6487	0.4278	6.7342	0.2375	11.9365	−3.3297
	318.15	0.1614	−0.9947	2.2917	0.3761	7.4173	0.2212	12.9854	−3.3045
Ethanol (1)+n-octane (2)	308.15	0.0644	1.5517	4.1135	13.1803	−0.8023	0.5694	0.3722	−2.9955
	318.15	0.0037	1.8760	3.4403	13.2823	−0.7895	0.4985	0.3384	−2.9824

$$\eta = \left[\sum_{i=1,2} x_i^2 \eta_i + 2H_{12} \prod_{i=1,2} x_i\right] \qquad (8)$$

Katti and Chaudhari [46,47]

$$\ln \eta V_m = \sum_{i=1,2} x_i \ln(\eta_i V_i) + x_1 x_2 (W_{vis}/RT) \qquad (9)$$

The following equation is used to calculate the standard deviations in percentage σ(%).

$$\sigma(\%) = \left[\frac{\sum\{(\eta_{exp.} - \eta_{theo.})/\eta_{exp.} \times 100\}^2}{(n-1)}\right]^{1/2} \qquad (10)$$

The notations used in above Eqs. (6)-(10) have their own significances and are well-defined in our previous paper [48].

The estimation capability of various correlations in terms of percentage standard deviation is given in Table 6. The Grunberg-Nis-

san (G-N) correlation gave the least value of percentage standard deviation, which is less than 8.8 percent for all studies binary system as seen from Table 6, while percentage standard deviation for other correlations was much larger, which means that Grunberg-Nissan (G-N) correlation is able to predict best η values among all other correlation for the present systems. Value of constant G_{12} in the Grunberg-Nissan (G-N) correlation is a measure of the strength of molecular interactions between the mixing components and is proportional to the interchange energy. Positive value of G_{12} indicates presence of strong interaction while negative value of G_{12} indicates the presence of weak interaction among the components of binary mixture. As we can observe from Table 6, the value of G_{12} is negative for all the studied binary mixtures, which means that there must be weak interaction among the components of binary mixture, which supports our experimental results.

CONCLUSIONS

The η for the systems 2-propanol (1)+n-alkanes (C_6, C_7, C_9) (2) at T=288.15 K to 303.15 K and ethanol (1)+n-alkanes (C_6, C_7, C_8) (2) at T=308.15 K to 318.15 K were experimentally measured. From these η values, $\Delta\eta$ values were calculated.Further, various correlations were applied to predict η values; the predicted data agree well with experimental η values, and it was found that Grunberg-Nissan correlation gave the best estimation for viscosity deviation. A theoretical approach suggested by Singh was applied to predict $\Delta\eta$ values. This approach yielded two constants, K^o and $K^\dagger$, which denote the unlike binary (1-2) interaction and degree of depolymerization of self-associated components, respectively. It was finalized that a modified approach [49] for associating combinations is able to give good estimation for the $\Delta\eta$ which can be observed in terms of standard deviation ($\sigma^\dagger$, σ^*) for existing systems.

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