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To cite this Article Saleh, Muhammad A. , Akhtar, Shamim and Ahmed, M. Shamsuddin (2004) 'Viscosity of aqueous solutions of some diamines', Physics and Chemistry of Liquids, 42: 2, 103-115 To link to this Article: DOI: 10.1080/0031910031000136219

URL: http://dx.doi.org/10.1080/0031910031000136219

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# VISCOSITY OF AQUEOUS SOLUTIONS OF SOME DIAMINES

## MUHAMMAD A. SALEH\*, SHAMIM AKHTAR and M. SHAMSUDDIN AHMED

Department of Chemistry, University of Chittagong, Chittagong – 4331, Bangladesh

(Received 8 February 2003)

Viscosities of the systems, ethylenediamine (ED) + water (W), 1,2-diaminopropane (DAP) + W, trimethylenediamine (TMD) + W and *N*,*N*-dimethyltrimethylenediamine (DMTMD) + W, have been measured for the whole range of composition at temperatures ranging from 303.15 to 323.15 K. The viscosities have been plotted against the mole fraction of diamines, showing some common characteristics for all the systems. On addition of diamines to water viscosities rise up very rapidly and pass through maxima. Following the maxima viscosities decline quite rapidly and monotonously up to about 0.6 mole fraction of diamines, which then fall down rather slowly. The hydrophobic and hydrophilic effects are considered responsible for the ascending part of viscosities in water-rich region, although the former is thought to be much more predominant than the latter. The height of viscosity maxima and their shifting to more water-rich region have been explained preferentially in terms of the hydrophobic effect. The declining part of viscosities is thought to be due to continuous destruction of cage structures formed by hydrophobic interaction. The excess viscosities have been calculated and plotted against mole fractions of diamines. The characteristics of excess viscosities have been explained by the same reasonings as used to explain viscosities.

*Keywords:* Viscosity; Excess viscosity; Hydrophobic hydration; Ethylenediamine; Trimethylenediamine; 1,2-diaminopropane; *N*,*N*-dimethyltrimethylenediamine

### **1. INTRODUCTION**

The present study is a part of our programme of studying the physico-chemical properties of aqueous solutions of hydrophobic organic solutes. In pursuance of this programme we reported in the recent past a number of studies on viscometric properties of aqueous solutions of some hydrophobic substances, such as alcohols [1], diols [2], acetone [3], dimethylsulfoxide, 1,4-dioxan, tetrahydrofuran [4], aliphatic amines [5], amides [6]. We noticed, to our surprise, that there is an absolute lack of viscometric data on aqueous solutions of diamines. This prompted us to undertake studies on aqueous solutions of diamines. As a first step towards this, we reported on the viscometric properties of the aqueous systems of ethylenediamine, trimethylenediamine and N,N-dimethyltrimethylenediamine, limited only to water-rich region [7]. The present study is an extended work of the above systems covering the whole range of concentration, with an additional system of 1,2-diaminopropane + water.

<sup>\*</sup>Corresponding author.

## 2. EXPERIMENTAL

The diamines under investigation were procured from the following manufacturers with quoted purities: ethylenediamine (Beijing Chemical Works, 99%), trimethylenediamine (Merk-Schuchardt, 98%), N,N-dimethyltrimethylenediamine (Merk-Schuchardt, 98%) and 1,2-diaminopropane (Merk-Schuchardt, >98%). The chemicals were used without further purification except that each of the diamines was kept over molecular sieves (4 A) for at least 2 weeks before its use. Thrice distilled water was used in the preparation of all the diamine solutions.

The density was measured by a 5 mL bicapillary pycnometer previously calibrated with distilled water. An Ostwald U-tube viscometer of the British standard Institution, with sufficiently long efflux time, was used so that no kinetic energy correction was necessary in viscosity measurement. The time of flow was recorded by a timer accurate up to  $\pm 0.01$  s. An analytical balance of accuracy  $\pm 0.0001$  g was used in density measurement. The solutions were prepared by mixing known masses of the components measured by the same balance. Mole fractions were accurate up to fourth place of decimal. For every measurement a thermostatic water bath controlled to  $\pm 0.05$  K was used. The maximum uncertainty in the measured viscosity was estimated to be  $\pm 0.3$  %.

### 3. RESULTS AND DISCUSSION

Viscosities of the aqueous solutions of ethylenediamine (ED), trimethylenediamine (TMD), 1,2-diaminopropane (DAP) and N,N-dimethyltrimethylenediamine (DMTMD) have been shown in Table I. The variation of viscosity as a function of mole fraction of the diamines,  $X_2$ , can be represented satisfactorily, as indicated by high values of  $r^2$ , by a polynomial equation of the type,

$$\eta = \sum_{i=0}^{n} A_i X_2^i \tag{1}$$

The values of the coefficients,  $A_i$  and  $r^2$  are shown in Table II.

Figures 1–4 represent the variation of viscosity as a function of mole fraction of ED, TMD, DAP and DMTMD, respectively, at temperatures ranging from 303.15 to 323.15 K. The viscosities of all the systems have been shown in Fig. 5 at 303.15 and 323.15 K for comparison. The viscosities, in the unit  $10^{-4}$  kg m<sup>-1</sup>s<sup>-1</sup>, of the pure liquids at 303.15 K are, ED (12.99), TMD (16.08), DAP (13.25) and DMTMD (12.998) and at 323.15 K, ED (9.04), TMD (10.896), DAP (9.091) and DMTMD (7.013).

The viscosity curves show the following general characteristics:

- (a) On addition of diamines to water, viscosities rise up very rapidly and attain maximum values.
- (b) Following the maxima, viscosities decline quite rapidly up to  $\sim 0.6$  mole fraction of the diamines, which then fall down rather slowly.
- (c) The temperature effect on viscosity is seen to be most prominent at compositions corresponding to maximum viscosities.
- (d) The positions of the maximum are found to occur at 0.331, 0.296, 0.299 and 0.252 mole fractions of diamines for ED + W, TMD + W, DAP + W and DMTMD + W, respectively, and virtually remain unaffected by change of temperature.

T (K)	303.15		308.15		313.15		318.15		323.15		
	η	$\eta^E$	η	$\eta^E$	η	$\eta^E$	η	$\eta^E$	η	$\eta^E$	
$X_{2}$ Water $(X_{1}) + ethylenediamine (X_{2})$											
0 0000	8 006	0.000	7 226	0.000	6 557	0 000	5 994	0.000	5 501	0.000	
0.1030	24 166	15 751	20 779	13 180	18 095	11 197	15 856	9 549	14 023	8 233	
0.2041	55 090	46 198	45 126	37.095	37 493	30 202	31 584	24 919	26 793	20.674	
0.2483	66 221	57 193	53 777	45.616	44 301	36.892	37 141	30.366	31 402	25.178	
0.2465	75 734	66 495	60.886	52 536	49.301	41 749	40.812	33 874	34 081	27 708	
0.2502	76 211	66 720	60.000	52.350	40.651	41.855	40.012	33.877	34 107	27.700	
0.3310	72 024	64 202	50.620	50.820	49.031	40.176	20 800	22 575	22 250	27.045	
0.4030	68 012	58 055	54 278	45 272	40.174	26 220	27 042	20 555	21 405	20.030	
0.4506	57 120	16 000	16 956	45.275	20 162	20.740	22 750	29.555	27.974	24.323	
0.5000	37.129 41.455	40.900	40.830	37.002	39.102	20.749	32.730	23.001	27.074	20.798	
0.3990	41.433	30./33	24.9/1	23.287	29.393	20.780	23.424	1/.300	22.003	14.392	
0.0989	29.098	1/.//8	24.914	14.008	21.725	12.407	19.023	10.499	10.803	8.904	
0.7936	21.4//	9.725	18.859	8.213	16.640	0.951	14.894	6.026	13.350	5.189	
0.8925	16.223	3.895	14.436	3.263	13.070	2.898	11.894	2.582	10.813	2.241	
1.0000	12.986	0.000	11.775	0.000	10.725	0.000	9.819	0.000	9.042	0.000	
0.0500	Water $(X_1)$ + trimethylenediamine $(X_2)$ 15 802 7 512 13 707 7 512 12 267 5 490 10 072 4 768 0 012 4 221										
0.0000	27 542	18 079	13.191	18 079	20.207	12 200	17.50	11 245	7.71J	9.612	
0.0900	42 602	24 720	25.555	24 720	20.290	13.200	17.000	10.260	10.400	9.012	
0.1438	43.002	52 (01	50.192	52 (01	41 002	23.004	25.000	19.200	22.223	10.140	
0.1990	02.889	55.091	50.881	33.091	41.882	34.301	33.339	28.487	29.300	23.204	
0.24/3	/5.680	00.107	60.548	00.10/	49.303	41.528	40.///	33.0/3	34.250	27.736	
0.2961	82.189	/2.34/	65.354	/2.346	53.135	45.094	43.783	36.437	36.764	30.029	
0.3465	79.913	69.719	64.011	69.719	52.199	43.874	43.358	35.753	36.320	29.349	
0.3974	74.378	63.815	60.215	63.815	49.463	40.841	41.200	33.324	34.701	27.483	
0.4955	58.778	47.467	48.319	47.467	40.379	31.154	34.143	25.718	29.346	21.628	
0.5995	43.070	30.908	36.440	30.908	31.266	21.356	26.737	17.688	23.137	14.850	
0.6969	32.236	19.220	27.792	19.220	24.190	13.592	21.137	11.462	18.658	9.801	
0.8006	25.009	11.016	21.774	11.016	19.208	7.825	17.118	6.729	15.341	5.833	
0.8936	19.971	5.041	17.616	5.041	15.727	3.591	14.165	3.090	12.814	2.682	
1.0000	16.080	0.000	14.447	0.000	13.059	0.000	11.914	0.000	10.896	0.000	
0.0000	20.026	01 (17	25 200	Water (	$(X_{I}) + 1,2$	-diaminop	ropane (X	<sup>(2)</sup>	16.050	10.470	
0.0998	30.036	21.617	25.288	17.692	21.54	14.645	18.549	12.248	16.250	10.470	
0.1997	65.963	57.110	52.584	44.598	42.867	35.617	35.591	28.966	29.770	23.688	
0.2522	77.022	67.931	60.951	52.752	49.129	41.685	40.408	33.607	33.647	27.403	
0.2992	80.426	71.117	63.825	55.431	51.386	43.764	42.106	35.143	34.955	28.562	
0.3412	77.928	68.420	61.961	53.388	50.329	42.544	41.103	33.992	34.525	27.995	
0.4012	70.954	61.154	56.777	47.943	46.9	38.198	38.227	30.899	32.044	25.315	
0.5022	55.384	45.073	45.038	35.745	37.343	28.901	31.397	23.689	26.711	19.631	
0.6011	39.212	28.374	32.885	23.120	27.92	19.048	23.948	15.848	20.796	13.356	
0.6913	28.441	17.099	24.431	14.215	21.132	11.848	18.498	10.024	16.321	8.536	
0.7912	21.189	9.261	18.542	7.802	16.339	6.576	14.504	5.595	13.025	4.839	
0.8998	16.888	4.289	15.012	3.672	13.116	2.805	12.162	2.755	11.012	2.367	
1.0000	13.251	0.000	11.924	0.000	10.844	0.000	9.891	0.000	9.091	0.000	
			Wate	$er(X_I) +$	N,N-dime	thyltrimeti	hylenedian	nine $(X_2)$			
0.0996	48.560	40.411	39.463	32.096	32.627	25.931	27.392	21.260	23.232	17.596	
0.1504	70.775	62.553	56.368	48.928	45.780	39.012	37.914	31.711	31.604	25.898	
0.1994	83.559	75.265	66.234	58.723	53.410	46.571	43.716	37.443	36.236	30.462	
0.2521	83.940	75.568	66.313	58.725	53.510	46.595	43.834	37.485	36.361	30.513	
0.2995	76.885	68.442	61.333	53.675	49.673	42.688	40.892	34.474	34.021	28.105	
0.3809	60.008	51.443	48.615	40.835	40.046	32.940	33.541	27.002	28.310	22.276	
0.5000	39.190	30.442	32.816	24.854	27.703	20.416	23.723	17.004	20.490	14.279	
0.5994	25.248	16.344	21.165	13.048	18.836	11.395	16.497	9.624	14.612	8.249	
0.6826	19.393	10.357	17.013	8.764	15.018	7.445	13.392	6.387	12.008	5.515	
0.7798	14.918	5.725	13.294	4.888	11.909	4.179	10.773	3.611	9.811	3.163	
0.8918	11.625	2.248	10.416	1.825	9.578	1.663	8.777	1.430	8.099	1.268	
1.0000	12.998	0.000	8.773	0.000	8.098	0.000	7.531	0.000	7.013	0.000	

TABLE I Experimental viscosities,  $\eta$ , and excess viscosities,  $\eta^E$  in  $10^{-4}$  kg m<sup>-1</sup> s<sup>-1</sup>, of aqueous diamines at different temperatures

System	Temperature (K)	$A_0$	$A_{I}$	$A_2$	$A_3$	$A_4$	$A_5$	$A_6$	$r^2$
	303.15	8.1957	-160.000	445.12	-16870	25913	-18165	4835.7	0.9988
	308.15	7.3391	-106.460	3393.8	-13125	20519	-14672	3995.2	0.9992
W + ED	313.15	6.0696	-61.648	2519.3	-9900.8	15591	-11219	3075.6	0.9996
	318.15	6.0181	-37.039	1954.3	-7807.5	12419	-9022.2	2497.5	0.9997
	323.15	5.5075	-17.020	1488.5	-6032.9	96357	-7019.3	1948.5	r <sup>2</sup> 0.9988 0.9992 0.9996 0.9997 0.9997 0.9997 0.9997 0.9983 0.9985 0.9990 0.9990 0.9990 0.9990 0.9999 0.9999 0.9999 0.9995 0.9999 0.9995 0.9995 0.9991 0.9946 0.9955 0.9961 0.9964
	303.15	8.9803	-105.21	4735.2	-19911	33458	-25585	7415.4	0.9976
	308.15	7.8524	-51.549	3425.6	-14585	24566	-18805	5456.5	0.9983
W+TMD	313.15	7.0004	-19.591	2531.5	-10909	18410	-14102	4094.6	0.9985
	318.15	6.2949	0.0825	1920.7	-8409.5	14257	-10946	3183.4	0.9990
	323.15	5.7506	12.276	1450	-6409.5	10836	-8277.7	2394.5	0.9990
	303.15	7.7986	-48.539	4449.7	-19570	33629	-26119	7664.7	0.9989
	308.15	7.0786	-10.869	3192.3	-14221	24467	-18988	5564.7	0.9990
W+DAP	313.15	6.4589	11.2	2324.1	-10470	17998	-13933	4074.7	0.9995
	318.15	5.8815	29.157	1680	-7760.7	13410	-10391	3036.2	0.9989
	323.15	5.4219	37.555	1233	5816.2	10062	-7778.1	2265.5	0.9991
	303.15	7.3653	383.28	1866	-15274	32661	-29034	9399.3	0.9946
	308.15	6.7559	317.47	1234.4	-10961	23682	-21114	6843.3	0.9951
W+DMTMD	313.15	6.201	267.49	799.6	-7934.5	17358	-15522	5033.6	0.9955
	318.15	5.7219	228.71	494.81	-5768.7	12826	-11521	3741.3	0.9961
	323.15	5.291	195.5	296.12	-4248.4	9611.6	-8675.2	2822.2	0.9964

TABLE II Coefficients,  $A_i$  of Eq. (1) expressing coefficient of viscosity,  $\eta \times 10^4/\text{kg m}^{-1}\text{s}^{-1}$ , and the square of the regression coefficient,  $r^2$ , for the systems

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FIGURE 1 Viscosity as a function of mole fraction of ethylenediamine, X<sub>2</sub> (ED), at different temperatures.



FIGURE 2 Viscosity as a function of mole fraction of trimethylenediamine,  $X_2$  (TMD), at different temperatures.



FIGURE 3 Viscosity as a function of mole fraction of 1,2-diaminopropane,  $X_2$  (DAP), at different temperatures.



X<sub>2</sub> ( DMTMD)

FIGURE 4 Viscosity as a function of mole fraction of N,N-dimethyltrimethylenediamine,  $X_2$  (DMTMD), at different temperatures.



FIGURE 5 Comparison of viscosities of different systems at 303.15 and 323.15 K against mole fractions of diamines  $(X_2)$ .

The viscosity curves of aqueous solutions of diamines are found to be typical of those of aqueous solutions of amphiphiles, such as, alcohols, amines, amides etc. [8–14]. The rapidly ascending part of the viscosity may be ascribed primarily to two effects: (i) hydrophilic effect and (ii) hydrophobic hydration. In the first, it is envisaged that diamines form complexes with water through hydrogen bonding, and in the second, the hydrocarbon moieties of the diamines are surrounded by water molecules forming cage-like structures, which are thought to be predominantly responsible for sharp rise of viscosity. The existence of unusually large negative excess molar volumes,  $V_m^E$ , and deep minima in water-rich region of these systems is inconsistent with this proposition [15]. With increased addition of diamines, more and more of such cages are formed, and hence the rapid rise of viscosity.

The maximum viscosity,  $\eta_{max}$  (in the unit  $10^{-4} \text{ kg m}^{-1} \text{ s}^{-1}$ ), for different systems at 303.15 K are found to be in the order, DMTMD + W (83.94) > TMD + W (82.19) > DAP + W (80.4) > ED + W (76.24). The  $\eta_{max}$  are considered to be extremely large compared to the viscosity of pure water and that their positions along the composition axis are different. These facts may conveniently be explained by the phenomenon of hydrophobic hydration. But before we proceed to do this we need to consider the structures of the diamines, which are shown below:

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To understand the degree of hydrophobic effect of the diamines, at least qualitatively, we can make use of the fact that CH<sub>3</sub> group is the most potent hydrophobic group, followed by CH<sub>2</sub> and CH groups, as formulated by Andini *et al.* [16]. Taking into consideration of this fact and perusal of the structures of these diamines above, we can easily arrive at the conclusion that, hydrophobic capacity of the diamines follows the order: DMTMD > TMD ~ DAP > ED. As the hydrophobicity is related to the extent of formation of cage structures, heights of  $\eta_{max}$  should follow the same order as above – which is indeed found to be the case.

It is to be noted that the positions of the  $\eta_{max}$  for different systems depend upon the degree of hydrophobicity in a manner that the greater the hydrophobicity the larger is the shifting of  $\eta_{max}$  towards more water-rich region. This is expected, because the greater the degree of hydrophobicity of diamines the larger is the number of water molecules to be required for the formation of cage structure, and hence, the shifting of  $\eta_{max}$  towards more water-rich region.

The effect of temperature on viscosity of the systems has been found to be highly significant, particularly on  $\eta_{\text{max}}$ . It is seen that, reduction of  $\eta_{\text{max}}$  for rise of temperature from 303.15 to 323.15 K,  $|\Delta \eta_{\text{max}}|$ , in the unit  $10^{-4} \text{ kg m}^{-1} \text{ s}^{-1}$ , has been found to be DMTMD + W (47.58) > TMD + W (45.43) ~ DAP + W (45.47) > ED + W (42.01). Such drastic reductions in  $\eta_{\text{max}}$  are expected, as water–water bonds formed by the cage structure are supposed to be less stable thermally than the normal water–water bonds [17,18]. The fragile nature of the water structure formed around the hydrocarbon residues of the diamines can be supported by large positive excess entropies of activation for viscous flow,  $\Delta S^{\#E}$ , of the solutions [15]. The above order of  $\eta_{\text{max}}$  can easily be justified by the hydrophobic capacity of the diamines.

When a reasonable number of cages are formed on addition of diamines to water, destruction of cages begins, probably through a process known as hydrophobic interaction [19]. However, as long as the rate of formation of cages is larger than the rate of destruction of cages, obviously, occurring in water-rich region, viscosity continues to increase. The maxima can be viewed as turning points when the two rates are equal. Following the maxima the rate of destruction becomes increasingly greater giving rise to the formation of more and more normal water structures and diamines, and hence the fairly rapid decrease of viscosity. The process is thought to continue until the pure state of diamines is reached.

The excess viscosities,  $\eta^E$ , of the systems were calculated by using the equation,

$$\eta^{E} = \eta - \exp(X_{1} \ln \eta_{1} + X_{2} \ln \eta_{2})$$
(2)

where  $\eta$  is the observed viscosity of the solution.  $X_1$  and  $X_2$  are the mole fractions of water and diamines respectively, and  $\eta_1$  and  $\eta_2$  are the corresponding viscosities of the pure components. The excess viscosities are shown in Table I at different temperatures. The values can be expressed satisfactorily by a Redlich–Kister polynomial equation of the form,

$$\eta^{E} = X_{1}X_{2}\sum_{i=0}^{n} A_{i}(1-2X_{1})^{i}$$
(3)

The coefficients,  $A_i$ , and standard deviations,  $\sigma$ , have been obtained by the least-squares method. The values of  $A_i$  and  $\sigma$  are listed in Table III. The  $\eta^E$  values have been plotted in Figs. 6–9 for different systems as a function of mole fractions of diamines at

System	Temperature (K)	$A_0$	$A_I$	$A_2$	$A_3$	$A_4$	$A_5$	σ	
W+ED	303.15	196.8861	-382.6084	48.8076	639.3310	-316.1813	-260.0907	1.00387	
	308.15	155.826	-286.4560	49.8241	429.5844	-255.9360	-133.4936	0.70746	<
	313.15	125.8152	-216.8419	39.8740	286.0087	-193.7383	-52.2017	0.40772	SL
	318.15	102.7861	-169.5302	36.5213	202.7203	-155.6187	-15.7798	0.30967	2
	323.15	85.1890	-133.9003	28.8246	145.0742	-120.3291	3.0652	0.26999	ISC
	303.15	188.9368	-387.6567	223.4220	520.8985	-478.5675	-151.048	1.73600	ΓY
	308.15	152.1279	292.2079	162.5013	365.7965	-349.2910	-89.8685	1.15482	0
W+TMD	313.15	124.4372	-224.9259	120.9057	265.0433	-260.2442	-55.9037	0.86502	T
	318.15	102.3049	-177.8599	95.9703	197.8406	-200.9278	-34.7398	0.58100	ő
	323.15	85.4952	-143.6976	73.3333	162.8661	-153.4328	-38.1218	0.45230	UE
	303.15	179.6054	-353.9753	245.3031	151.1257	-470.0440	335.6977	0.60336	ОО
	308.15	142.8805	-268.7546	180.2639	110.7420	-341.1065	246.5402	0.46127	S
W + DAP	313.15	115.4734	-207.9103	135.5855	82.4100	-256.6871	178.6314	0.30732	OS OS
	318.15	78.4543	-128.8654	77.5609	34.0834	-138.2045	124.1244	0.16328	Ľ
	323.15	94.4765	-160.3346	101.6875	37.2824	-185.1873	172.3875	0.22839	ILL
	303.15	114.0240	-296.6553	556.8312	-503.5781	-552.0907	861.1757	1.44165	g
W+DMTMD	308.15	92.5602	-235.7194	418.3454	-345.5874	-406.0019	598.0644	1.23259	S
	313.15	76.9642	-183.7923	314.9245	-277.3597	-295.3845	467.7151	0.88246	
	318.15	64.3437	-148.5349	242.5957	-207.4651	-219.6091	346.7598	0.69654	
	323.15	54.1982	-120.3688	189.3426	-160.9532	-165.5002	265.3007	0.56034	

TABLE III Coefficients,  $A_i$ , of Redlich-Kister equation, Eq. (3), expressing excess viscosity,  $\eta^E$ , and standard deviation,  $\sigma$ , in  $10^{-4}$  kg m<sup>-1</sup> s<sup>-1</sup> for the systems

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FIGURE 6 Excess viscosity of the system water  $(X_1)$  + ethylinediamine  $(X_2)$  at different temperatures.



FIGURE 7 Excess viscosity of the system water  $(X_1)$  + trimethylenediamine  $(X_2)$  at different temperatures.



FIGURE 8 Excess viscosity of the system water  $(X_1) + 1,2$ -diaminopropane  $(X_2)$  at different temperatures.



FIGURE 9 Excess viscosity of the system water  $(X_1) + N,N$ -dimethyltrimethylenediamine  $(X_2)$  at different temperatures.



FIGURE 10 Comparison of excess viscosity of different systems against mole fractions of diamines  $(X_2)$  at 303.15 and 323.15 K.

different temperatures. Figure 10 shows the comparison of  $\eta^E vs. X_2$  curves at 303.15 and 323.15 K for all the systems.

The excess viscosities have been found to be positive and large in magnitude, indicating that the solutions are highly nonideal. The characteristics of the  $\eta^E$  curves are very much similar to those of  $\eta$  curves as described earlier. The excess viscosities can be explained by the same reasonings as those put forward for the explanation of viscosities.

#### Acknowledgement

The authors gratefully acknowledge the financial support of the Ministry of Science, Information and Communication Technology, Govt. of the People's Republic of Bangladesh, as a special allocation for the project, "Physical Properties and Molecular Interactions in Liquid Systems".

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