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Excess Molar Volumes and Kinematic Viscosities of Diethylene Glycol + Dimethyl Sulfoxide at T = (298.15, 303.15, 308.15, 313.15 and 318.15) K

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Densities (ρ) and kinematic viscosities (ν) for the binary system of diethylene glycol + dimethyl sulfoxide were measured at T = (298.15, 303.15, 308.15, 313.15 and 318.15) K. The density values, which were determined using a capillary pycnometer, were used to calculate the excess molar volumes (V_m^E). The kinematic viscosity (ν) data measured with an Ubbelohde capillary viscometer were used to calculate activation enthalpy (ΔH^*) and activation entropy (ΔS^*) for the viscous flow.

Key Words: Excess molar volume, Viscosity, Activation enthalpy, Activation entropy.

INTRODUCTION

Sulfur dioxide produce from the burning of fossil fuels with high sulfur content, is an important air pollutant. The large amount of SO_2 emission is responsible for acid rain, which leads to economic loss per year[1]. At the same time, SO_2 can bring harm to humans and the environment. Accordingly, the removal of SO_2 from industrial flue gas becomes one of the most serious problems.

Among all of the methods ever have developed on the desulfurization, wet desulfurization methods[2-4] and especially wet limestone method, were always the first choice for both China and most of the western developed countries. But some disadvantages, such as huge magnitude of capital investment, high operating cost and secondary pollution, still blocked its development. Nowadays, organic solvents, which was used as absorbents, have been identified as an option among the regenerative processes[5-9], which attracting more and more interests. Of the numerous organic solvents, diethylene glycol (DEG) has been introduced because of its low vapour pressure, low toxicity, low melting point and high chemical stability. In addition, dimethyl sulfoxide (DMSO), being a highly polar solvent, can increase the speed of absorption when added to diethylene glycol as an optimum media.

Incomplete the values of basic data for pure solvents and binary systems in the current literatures encourages us to carry out a series of systematic measurements of density (ρ) and viscosity (η) over the whole concentration range at T = (298.15 to 318.15) K with a step of 5 K. Meanwhile, the excess molar volume (V_m^E), viscosity deviation (Δη), activation enthalpy (ΔH^*) and activation entropy (ΔS^*) were calculated. The peculiarity of this work is used to provide important basic data for the design and operation of the absorption and desorption processes for removal of SO_2 with potential industrial application of the solutions containing diethylene glycol.

EXPERIMENTAL

The analytical grades diethylene glycol (DEG) with a purity of 99 % (mass fraction) and DMSO with a purity of 99.8 % (mass fraction) were purchased from Beijing Reagent Company (Beijing, China). All the samples were kept over 4 Å molecular sieves and decompression filtration to reduce the water content prior to each experiment before used. Binary mixtures were prepared by mass on a digital electronic balance, which has a stated repeatability of ± 1×10^{-4} gram. Solutions were kept in airtight sample bottles and closed with caps to avoid the effects of atmospheric humidity. Then for each ratio, a series of solutions of different concentrations were made. Distilled water and high-performance liquid chromatography (HPLC) grade ethanol were used.

Density and viscosity measurements: Density (ρ) of the DEG and DEG + DMSO mixture solutions were measured within a precision of ± 0.0001 g cm^{-3} using a capillary-type pycnometer of 10 cm^3 volume. Doubly distilled, deionized and degassed water were used to calibrate the pycnometer. The experiments, including density and viscosity mentioned in the following, was carried out at T = (298.15 to 318.15) K with a step of 5 K. Each experimental density value was an average of at least three measurements. The purity of DEG was checked by measuring and comparing the densities with corresponding literature values[10-13].

Kinematic viscosities (ν) were measured by a calibrated Ubbelohde type viscometer with a capillary diameter of 0.90 mm. The viscometer was calibrated with high pure water and ethanol (HPLC grade) in the experimental temperature range. The thoroughly cleaned and perfectly dried viscometers kept in a thermostatically controlled and well-stirred water bath for (20 to 25) min to attain thermal equilibrium. An electronic digital stop-watch with a readability of 0.01 s was used for the flow time measurements. The averaged of 18 sets of flow times for each fluid was taken for the purpose of the calculation of viscosity from the equation:

$$\nu = At - \frac{B}{t} \tag{1}$$

where ν is the kinematic viscosity, A and B are viscometer constant and t is the flow time in seconds, respectively. The viscosity of DEG was checked by measuring and comparing with corresponding literature values[12-15].

Absolute viscosity can be obtained by multiplying the determined kinematic viscosity by the measured density ($\eta = \nu \times \rho$). For the aims of this paper, the kinematic viscosity (ν) was chosen rather than the absolute viscosity (η) since it is not affected by experimental errors associated with density measurements. The thermodynamic parameters of viscous flow, such as activation free energy (ΔG^*), activation enthalpy (ΔH^*) and activation entropy (ΔS^*), of the binary mixtures were evaluated on the basis of Eyring's approach to Andrade's theory[16].

RESULTS AND DISCUSSION

Excess molar volume: A comparison of measured density values in the literature data[12-16] and that of the calculated values has showed in Table-1, the results show that a good agreement was found between the present measurements of density and viscosity with the data in the literatures.

TABLE-1
COMPARISON OF EXPERIMENTAL DENSITIES (ρ), VISCOSITIES (η) OF DEG WITH LITERATURE VALUES AT VARIOUS TEMPERATURES

T (K)	ρ (g cm^{-3})		η (mPa s)	
	Expt.	Lit.	Expt.	Lit.
298.15	1.1128	1.1129[12], 1.1131[15]	17.0	27.93[16]
303.15	1.1098	1.10948[15], 1.1099[14]	12.4	22.09[17], 21.75[18]
308.15	1.1051	..10557[13], 1.10578[12]	10.4	17.36[18]
313.15	1.0998	1.10211[15]	8.19	12.30[19]
318.15	1.0963	1.09835[14]	7.32	12.19[18]

Experimental densities under atmospheric for the binary solution of DEG + DMSO over a temperature range from (298.15 to 318.15) K at intervals of 5 K throughout the whole concentration range were presented in Table-2. The values of the measured density at different temperature are shown in Fig. 1.

TABLE-2
EXPERIMENTAL DENSITIES (ρ) OF DEG (1) + DMSO (2) AT DIFFERENT TEMPERATURE

	ρ (g cm^{-3})				
	Temperature (K)				
	298.15	303.15	308.15	313.15	318.15
1.0000	1.1128	1.1093	1.1052	1.0998	1.0963
0.9333	1.1125	1.1078	1.1037	1.0992	1.0957
0.8689	1.1122	1.1076	1.1030	1.0985	1.0952
0.8066	1.1120	1.1071	1.1023	1.0979	1.0946
0.7465	1.1120	1.1066	1.1023	1.0973	1.0937
0.6883	1.1106	1.1060	1.1015	1.0966	1.0927
0.6320	1.1102	1.1050	1.1005	1.0958	1.0919
0.5775	1.1099	1.1044	1.0998	1.0950	1.0910
0.5248	1.1090	1.1030	1.0986	1.0939	1.0901
0.4736	1.1085	1.1025	1.0979	1.0931	1.0894
0.4240	1.1074	1.1016	1.0966	1.0917	1.0880
0.3759	1.1062	1.1014	1.0963	1.0916	1.0878
0.3292	1.1052	1.0995	1.0943	1.0896	1.0856
0.2839	1.1043	1.0983	1.0933	1.0881	1.0847
0.2398	1.1033	1.0972	1.0919	1.0869	1.0827
0.1970	1.1017	1.0959	1.0907	1.0855	1.0813
0.1554	1.1008	1.0947	1.0898	1.0842	1.0797
0.1150	1.0994	1.0933	1.0884	1.0827	1.0779
0.0756	1.0979	1.0919	1.0866	1.0806	1.0762
0.0373	1.0965	1.0905	1.0847	1.0791	1.0744
0.0000	1.0946	1.0888	1.0831	1.0785	1.0725

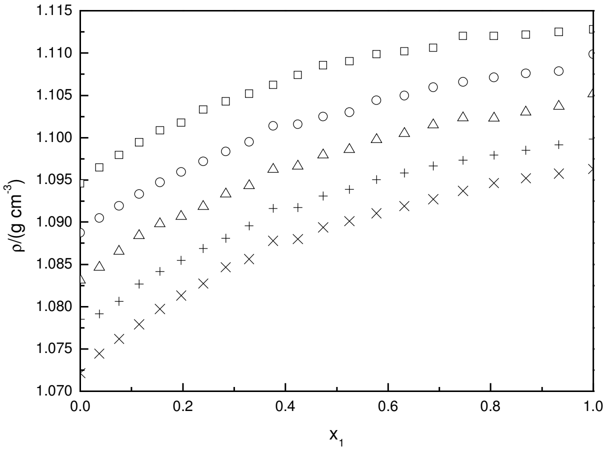

Fig. 1. Experimental density with mole fraction for DEG (1) + DMSO (2): □ 298.15 K; ○ 303.15 K; △ 308.15 K; + 313.15 K and × 318.15 K

Experimental viscosities under the same condition were presented in Table-3. Fig. 2 shows the solutions viscosities of DEG in DMSO at different temperatures.

Conclusions can be drawn from Fig. 1 and Table-2 that the density values increase with the increasing DEG concentration in binary solution over the whole concentration range. But the density values decreased with the increment of temperature at the same concentration. Meanwhile, in all cases, Fig. 2 together with Table-3 indicate that experimentally measured viscosities of the binary solutions of DEG + DMSO at T = (298.15, 303.15, 308.15, 313.15 and 318.15) K increase with the increasing DEG concentration and decrease with the increasing temperatures.

The excess molar volume (V_m^E) was calculated based on the measured density according to the eqn. 2:

$$V_m^E = \frac{x_1 M_1 + x_2 M_2}{\rho_m} - \left(x_1 \frac{M_1}{\rho_1} + x_2 \frac{M_2}{\rho_2} \right) \tag{2}$$

TABLE-3
EXPERIMENTAL VISCOSITIES (ρ) OF DEG + DMSO AT DIFFERENT TEMPERATURE

	η (mPa s)				
	Temperature (K)				
	298.15	303.15	308.15	313.15	318.15
1.0000	1.40	1.21	1.12	1.00	0.954
0.9333	1.54	1.32	1.22	1.09	1.040
0.8689	1.71	1.44	1.34	1.18	1.130
0.8066	1.88	1.58	1.47	1.29	1.240
0.7465	2.10	1.74	1.62	1.42	1.350
0.6883	2.35	1.94	1.79	1.55	1.490
0.6320	2.64	2.16	1.99	1.71	1.640
0.5775	2.97	2.41	2.21	1.89	1.800
0.5248	3.34	2.68	2.44	2.09	1.990
0.4736	3.78	3.31	2.98	2.51	2.210
0.4240	4.26	3.38	3.05	2.56	2.440
0.3759	4.84	3.81	3.41	2.86	2.700
0.3292	6.10	4.29	3.82	3.17	2.990
0.2839	6.23	4.86	4.29	3.54	3.300
0.2398	7.13	5.50	4.83	3.98	3.670
0.1970	8.16	6.27	5.48	4.47	4.150
0.1554	9.43	7.13	6.17	5.01	4.560
0.1150	10.8	8.11	6.97	5.61	5.120
0.0756	12.5	9.31	7.91	6.34	5.750
0.0373	14.4	10.7	9.03	7.12	6.450
0.0000	17.0	12.4	10.4	8.19	7.320

TABLE-4
EXCESS MOLAR VOLUMES (V_m^E) OF DEG + DMSO AT DIFFERENT TEMPERATURE

	V_m^E (cm^{-3} mol^{-1})				
	Temperature (K)				
	298.15	303.15	308.15	313.15	318.15
1.0000	0.000	0.000	0.000	0.000	0.000
0.9333	-0.052	0.081	0.030	-0.035	-0.058
0.8689	-0.101	0.009	-0.007	-0.071	-0.114
0.8066	-0.162	-0.039	-0.039	-0.111	-0.169
0.7465	-0.233	-0.082	-0.133	-0.148	-0.192
0.6883	-0.192	-0.115	-0.155	-0.180	-0.209
0.6320	-0.227	-0.120	-0.158	-0.198	-0.239
0.5775	-0.269	-0.158	-0.188	-0.217	-0.264
0.5248	-0.272	-0.129	-0.178	-0.208	-0.281
0.4736	-0.300	-0.168	-0.212	-0.227	-0.315
0.4240	-0.279	-0.177	-0.193	-0.201	-0.298
0.3759	-0.257	-0.237	-0.245	-0.270	-0.369
0.3292	-0.244	-0.172	-0.180	-0.193	-0.294
0.2839	-0.240	-0.163	-0.187	-0.162	-0.308
0.2398	-0.234	-0.154	-0.158	-0.148	-0.250
0.1970	-0.184	-0.138	-0.148	-0.122	-0.233
0.1554	-0.182	-0.124	-0.163	-0.102	-0.202
0.1150	-0.145	-0.098	-0.141	-0.072	-0.156
0.0756	-0.105	-0.074	-0.086	-0.002	-0.117
0.0373	-0.067	-0.046	-0.030	0.028	-0.077
0.0000	0.000	0.000	0.000	0.000	0.000

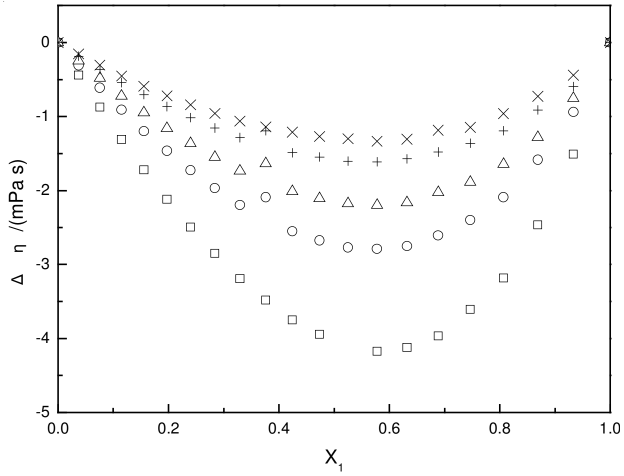

Fig. 2. Experimental viscosities with mole fraction for DEG (1) + DMSO (2): □ 298.15 K; o 303.15 K; Δ 308.15 K; + 313.15 K and × 318.15 K

where ρ_m is the density of the mixture and x_1, ρ_1, M_1, x_2, ρ_2 and M_2 are the mole fractions, densities and molecular weights of the pure DEG and pure DMSO, respectively. The results of V_m^E are listed in Table-4 and the dependence of V_m^E at various temperatures is displayed in Fig. 3.

Fig. 3 shows that the excess molar volumes are negative for all the mixtures over the entire range of composition. The value became less negative with the increasing temperature and arrived the minimum around molar fraction 0.40 for DEG at all temperatures. Additionally, these less negative V_m^E values indicate that there is a maximum volume contraction on mixing at a rate of 2:3. That means two DEG molecules can bond three DMSO molecules to inform a stable structure. Consequently, this kind of interaction is affected by temperature and composition, which can be explained as the molecules becoming more active with the increasing temperatures. So DEG can interact with DMSO more easily.

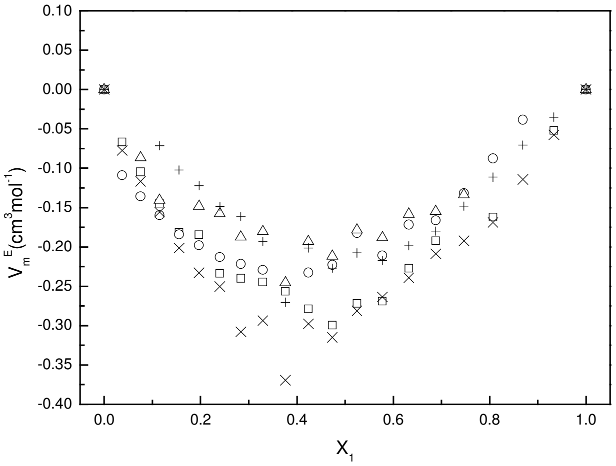

Fig. 3. Excess molar volumes with mole fraction for DEG (1) + DMSO (2): □ 298.15 K; o 303.15 K; Δ 308.15 K; + 313.15 K and × 318.15 K

The calculated V_m^E values were correlated using Redlich-Kister-type polynomial eqn. 3:

$$V_m^E\,(cm^3mol^{-1}) = x_1x_2\sum_{i=0}^{n} A_i(2x_1-1)^i \quad (3)$$

where A_i are the parameters of the Redlich-Kister polynomial equation, which were obtained using the method of least squares and x_1 is the mole fraction of DEG and x_2 is the mole fraction of DMSO. The parameters of the Redlich-Kister equation representing the excess molar volumes for the DEG and DEG + DMSO systems are presented in Table-5 together with the corresponding standard deviation, which was estimated using the following eqn. 4:

$$\sigma_{V_m^E} = \left[\sum (V_{calc}^E - V_m^E)^2/(N-m)\right]^{1/2} \quad (4)$$

where N is the total number of experimental points and m is the number of A_i coefficients considered.

TABLE-5
COEFFICIENTS AND STANDARD DEVIATIONS
OF V_m^E FOR DEG + DMSO

T (K)	A_0	A_1	A_2	A_3	A_4	σ (cm^{-3} mol^{-1})
298.15	-0.885	0.211	-0.069	-0.486	1.145	0.0193
303.15	-0.912	0.276	-0.184	1.047	0.182	0.0229
308.15	-0.803	0.118	-0.049	1.160	0.763	0.0194
313.15	-0.885	0.211	-0.068	-0.486	1.145	0.0193
318.15	-1.215	0.622	-0.024	-0.531	-0.320	0.0180

The viscosity values η ($\eta = \nu \times \rho$) for the various mixtures were used to calculate the viscosity deviation Δη, defined by eqn. 5:

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{5}$$

where η is the viscosity of the mixture, η_1 and η_2 are the viscosities of pure DEG and pure DMSO and x_1 is the mole fraction of DEG and x_2 is the mole fraction of DMSO, respectively. The results of the viscosity deviation are listed in Table-6 and the dependence of Δη is plotted in Fig. 4.

TABLE-6
VISCOSITY DEVIATIONS (Δη) OF DEG (1) +
DMSO (2) AT DIFFERENT TEMPERATURE

	Δη (mPa s)				
	Temperature (K)				
	298.15	303.15	308.15	313.15	318.15
1.0000	0.000	0.000	0.000	0.000	0.000
0.9333	-0.440	-0.306	-0.248	-0.186	-0.155
0.8689	-0.873	-0.610	-0.483	-0.364	-0.306
0.8066	-1.310	-0.909	-0.723	-0.540	-0.450
0.7465	-1.720	-1.200	-0.948	-0.704	-0.592
0.6883	-2.120	-1.460	-1.160	-0.866	-0.719
0.6320	-2. 500	-1.730	-1.360	-1.020	-0.840
0.5775	-2.850	-1.970	-1.550	-1.160	-0.957
0.5248	-3.190	-2.200	-1.740	-1.280	-1.060
0.4736	-3.480	-2.090	-1.640	-1.190	-1.140
0.4240	-3.750	-2.550	-2.010	-1.490	-1.210
0.3759	-3.940	-2.680	-2.110	-1.550	-1.270
0.3292	-3.480	-2.770	-2.170	-1.600	-1.300
0.2839	-4.170	-2.790	-2.200	-1.610	-1.330
0.2398	-4.120	-2.750	-2.160	-1.570	-1.310
0.1970	-3.970	-2.610	-2.030	-1.480	-1.190
0.1554	-3.610	-2.400	-1.880	-1.360	-1.150
0.1150	-3.180	-2.090	-1.640	-1.190	-0.962
0.0756	-2.460	-1.590	-1.280	-0.911	-0.726
0.0373	-1.510	-0.938	-0.752	-0.592	-0.442
0.0000	0.000	0.000	0.000	0.000	0.000

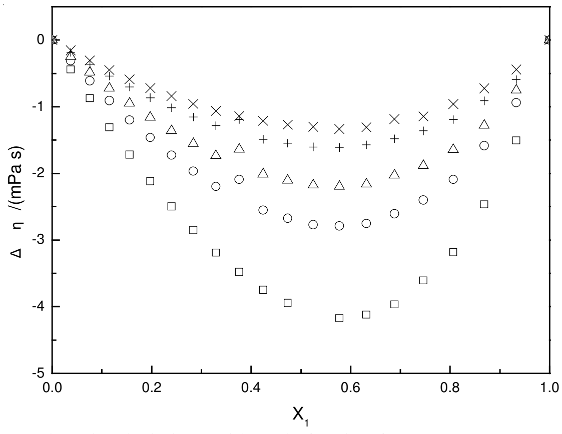

Fig. 4. Viscosity deviations with mole fraction for DEG (1) + DMSO (2): □ 298.15 K; o 303.15 K; Δ 308.15 K; + 313.15 K and × 318.15 K

Fig. 4 shows that in all cases the Δη values are negative over the whole composition range for mixtures. The viscosity deviations decrease with the increasing temperatures. The deviations, Δη, were also represented by the Redlich-Kister eqn. 6. The coefficients B_i and the standard deviation are presented in Table-7.

$$\Delta\eta/\mathrm{mPa\cdot s} = x_1x_2\sum B_i(2x_1-1)^i \tag{6}$$

TABLE-7
COEFFICIENTS AND STANDARD DEVIATIONS
OF VISCOSITY (Δη) FOR DEG + DMSO

T (K)	B_0	B_1	B_2	B_3	B_4	σ (mPa s^{-1})
298.15	-15.6	-4.92	-4.89	-2.01	2.80	0.136
303.15	-10.7	-3.62	-1.15	0.58	-0.31	0.068
308.15	-8.45	-2.71	-0.78	0.072	-0.87	0.057
313.15	-6.21	-1.76	-0.17	-0.17	-1.39	0.048
318.15	-5.17	-1.26	-0.34	-0.18	-0.35	0.015

Thermodynamic parameters: The structural changes occurred as molecular interactions in mixed solvents can be used to explain excess molar volumes, it can also be further investigated by thermodynamic analysis of the viscosity. Therefore, the thermodynamic parameters of activation of viscous flow, such as free energy (ΔG^*), enthalpy (ΔH^*) and entropy (ΔS^*), of the binary mixtures have been evaluated with the viscosity expressed in the eqns. 7 and 8:

$$\upsilon = \frac{hN_A}{\langle M\rangle}\exp\left(\frac{\Delta G^*}{RT}\right) \tag{7}$$

$$\langle M\rangle = \sum_i X_i M_i \tag{8}$$

where <M> is the average molar mass, h is Planck's constant, N_A is the Avogadro number, R is the gas constant and T is the absolute temperature. Eqn. 7 can also been write in eqn. 10 by applying the standard thermodynamic eqn. 9, where ΔH^* and ΔS^* represent the active enthalpy and active entropy for the viscosity low, respectively.

$$\Delta G^* = \Delta H^* - T\cdot\Delta S^* \tag{9}$$

$$R * \ln\frac{\upsilon^*\langle M\rangle}{h * Na} = \frac{\Delta H^*}{T} - T\cdot\Delta S^* \tag{10}$$

The non-linearity of the plots of $R\ln(\nu\langle M\rangle/hNa)$ against 1/T in Fig. 5 deduce the conclusion that slop value is equal to activation enthalpy and the intercept represents the activation entropy in negative. Conclusions can also been drawn from Fig. 5 that ΔH^* values are all positive while the ΔS^* values are generally negative. $\Delta H^* > 0$ and decrease as the temperature increase because interaction occurrence more easily as molecular becoming more active. Together with the V_m^E values become more negative with the increasing temperature can introduce the conclusion that interaction of DEG with DMSO is stronger than that of DEG with DEG. On the contrary, ΔS^* values become more negative with the increasing temperature as presented in Table-8. In this system ΔS^* appears to increase with the mole fraction increase and arrive at maxima when it was one. There is one reason that the formation of complexes in DEG + DMSO system increases the orderliness. To sum up,

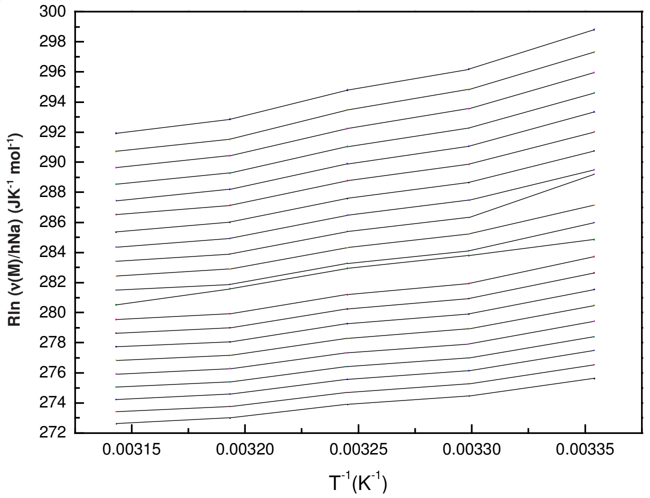

Fig. 5. Plots of Rln(ν(M)/hNa) against 1/T for DEG (1) + DMSO (2) system at various temperatures

TABLE-8
ACTIVATION ENTHALPY (ΔH^*) AND ACTIVATION ENTROPY (ΔS^*) OF DEG + DMSO

	10^3 ΔH^* (J mol^{-1})	ΔS^* (J K^{-1} mol^{-1})
1.0000	32.54 ± 2.68	-189.27 ± 8.72
0.9333	31.30 ± 2.64	-191.96 ± 8.56
0.8689	29.94 ± 2.57	-195.15 ± 8.34
0.8066	28.76 ± 2.54	-197.75 ± 8.25
0.7465	27.92 ± 2.40	-199.32 ± 7.81
0.6883	26.14 ± 2.45	-203.98 ± 7.96
0.6320	25.53 ± 2.34	-204.78 ± 7.59
0.5775	24.47 ± 2.30	-207.09 ± 7.49
0.5248	26.69 ± 4.15	-198.97 ± 13.49
0.4736	22.34 ± 2.36	-211.86 ± 7.67
0.4240	21.34 ± 2.40	-214.06 ± 7.80
0.3759	20.61 ±0.96	-215.81 ±3.11
0.3292	19.74 ± 2.26	-217.18 ± 7.35
0.2839	18.94 ± 2.17	-218.79 ± 7.06
0.2398	18.01 ± 2.13	-220.82 ± 6.91
0.1970	17.25 ± 1.96	-222.32 ± 6.37
0.1554	16.51 ± 1.86	-223.77 ± 6.04
0.1150	15.75 ± 1.72	225.32 ± 5.58
0.0756	15.38 ± 1.58	-225.67 ± 5.13
0.0373	14.78 ± 1.44	-226.73 ± 4.68
0.0000	14.23 ± 1.25	-227.67 ± 4.05

all the above analysis derived the result that interaction between DEG and DMSO is more easily than DEG and DEG. Obviously this result is in good agreement with the theoretical approach.

Conclusion

Densities and viscosity for the binary systems of DEG + DMSO were measured over the temperature range (298.15 to 318.15) K. The densities and viscosities values increase with increasing mole fraction of DEG and are decrease with increasing temperature. The experimental density values were then used for the calculation of excess molar volumes V_m^E, which were negative at all compositions and temperatures. Kinematic viscosities (ν) obtained from the measurement were used to deduce the activation enthalpy and activation entropy, showing the movement tendency in the system, which certified the interaction in dynamic view. While the viscosity deviations $\Delta\eta$ are also negative over the whole composition range and their values become less negative as the temperature increase. Since the binary mixtures could be used as a hybrid solvent for the capture of SO_2 from flue gases, the present data on density, viscosity and excess molar volumes will certainly be useful for the efficient design of gas absorbed for commercial applications in industry.

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