Excess Thermodynamic Properties of Binary Liquid Mixtures of Dimethyl Carbonate with Xylene

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Abstract The experimental data of density and viscosity of the binary liquid mixtures of Dimethyl Carbonate (DMC) with *o*-xylene and *m*-xylene have been explored over the entire composition range at atmospheric pressure and four different temperatures (298.15 K, 303.15 K, 308.15 K & 313.15 K) to calculate excess molar volume (V^E), deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation (G^E). The interaction parameters have been estimated by correlating the experimental viscosity data using the McAllister equation. The experimentally determined values of V^E, $\Delta\eta$, and G^E have been fitted to the Redlich-Kister polynomial equation to calculate coefficients and standard error values. The variation in thermodynamic properties with the composition and temperature of the binary mixtures has been discussed in terms of intermolecular interactions. The density of both binary liquid mixtures, i.e., DMC + *o*-xylene and DMC + *m*-xylene, decreases with an increase in temperature and increases as the mole fraction of DMC increases. Still, viscosity decreases with an increase in temperature and an increase in the mole fraction of DMC in both mixtures. This paper will help the reader to improve their practical skills, understanding of chemical thermodynamics principles, and ability to interpret experimental data and identify trends.

Keywords: *liquid mixture, excess thermodynamic properties, dimethyl carbonate, xylene, temperature*

1. Introduction

The understanding of thermodynamic and transport properties of liquid-liquid mixtures is essential for their application in chemical, textile, leather, and nuclear industries. When two liquids are mixed, the resulting changes in physical and thermodynamic properties can be considered as a sum of several contributions due to free volume change, change in energy, change in molecular orientations, and steric hindrances. The mixing of different compounds gives rise to solutions that generally do not behave as ideal solutions. The deviation from ideal behaviour is expressed by many thermodynamic variables, particularly by excess properties. Excess properties are defined as the difference between the actual property value of a real solution and the value it would have as an ideal solution at the same temperature, pressure, and composition. Equation 1 gives the excess properties of a mixture related to those of the pure substances in an ideal mixture.

$$Z^E = Z_m - \sum x_i Z_j^{id} \qquad (1)$$

Where Z corresponds to the specific property under consideration, Z^E is the excess molar property, Z^m is the mixture property, Z^{id} is the property of the ideal mixture, and *x* is the mole fraction. The measurement of density and viscosity in liquid mixtures and solutions has also been found to be an important tool for studying the physicochemical properties of the mixtures and solutions. The density of a material is defined as its mass per unit volume. It is generally represented as ρ(rho). The density of all the fluids depends on the temperature and pressure; the variation in density with changes in these variables may be small or large. Increasing the pressure on an object decreases its volume and therefore increases its density. Increasing the temperature of a substance (with some exceptions) decreases its density by increasing the volume of that substance [1]. Viscosity is a function of temperature and pressure and has a direct and large effect on heat transfer coefficients [2]. Viscosity is expressed in two distinct ways: Absolute or dynamic viscosity and Kinematic viscosity. Dynamic viscosity is the tangential force per unit area required to slide one layer against another layer, whereas Kinematic viscosity is the ratio of the absolute viscosity to the density of a fluid ($\nu = \eta/\rho$), where ν and η represent kinematic viscosity and absolute viscosity, respectively.

In this work, binary liquid mixtures of DMC with *o*-xylene and *m*-xylene have been prepared, and excess thermodynamic properties have been studied at different temperatures and fitted to the Redlich-Kister polynomial equation to estimate the standard deviation values.

2. Literature Review

Various research papers have been reported on the excess properties of 1-iodobutane with benzene, toluene, *m*-xylene, and mesitylene [3], 2-butanone with *o*-xylene, *m*-xylene, *p*-xylene, and isopropyl benzene [4], 1,4-dioxane + benzene, positive for 1,4-dioxane + *o*-xylene, + *m*-xylene, + *p*- xylene, and + mesitylene [5], methyl acrylate (MA) with benzene, toluene, *o*-xylene, *m*-xylene, *p*-xylene, and mesitylene [6], 3- Pentanone + Ethyl benzene and 3-Pentanone + *o*-xylene [7], 2-Propanol and 3-Pentanone with *p*-xylene [8], acetonitrile

with benzene, toluene, *m*-xylene and mesitylene [9], DMC with 2-propanol up to 2-heptanol [10], DMC, *n*- octane, *n*-decane [11], DMC + 1-hexanol/1-octanol [12] and DMC + methanol, or + ethanol, or +1-propanol [13].

It has been observed from the available literature that several studies have been reported with different combinations of xylenes and other chemicals used at different temperatures to study their excess properties, but studies involving DMC are rare. It is an environment-friendly chemical that belongs to the organic carbonate family and is used as a replacement for various hazardous chemicals [14]. It is an excellent additive for fuels. a strong contender to help the refining industry meet the Clean Air Act because DMC has about three times the oxygen content per volume as methyl *tert*-butyl ether (MTBE) [15]. Also, Xylene is an aromatic hydrocarbon widely used in industry and medical technology as a solvent in the printing, rubber, paint, and leather industries [16-18]. Keeping this in view, a study has been undertaken to study the excess thermodynamic properties of binary liquid mixtures of DMC with o-xylene and m-xylene at different temperatures.

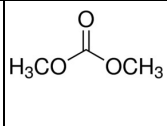
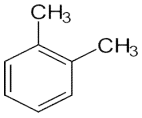
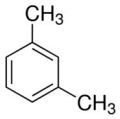
3. Experiment

The density and viscosity of the prepared binary mixtures were measured to study their excess properties for the entire composition range at atmospheric pressure and various temperatures.

3.1. Materials

DMC, o-Xylene, and m-Xylene of AR grade were purchased from S.R.L. Pvt. Ltd., Mumbai and stored over molecular sieves to remove the moisture present. Table 1 presents the properties of the materials used.

Table 1. Properties of materials[3-7]

Properties	Material		
	DMC	o-Xylene	m-Xylene
Chemical formula	$C_3H_6O_3$	C_8H_{10}	C_8H_{10}
Structure			
Molar mass (g mol^{-1})	90	106.16	106.16
Density (g/ml)	1.069 – 1.073	0.88	0.86
Viscosity (mPa.s) (at 25 ºC))	0.585	0.810	0.620
Melting point (ºC)	2-4	-24	-48
Boiling point (ºC)	90	144.4	139
Solubility in water	13.9 g/100ml	Insoluble	Insoluble

3.2. Measurement

The purity of the chemicals was checked by comparing the values of densities and viscosities with the reported literature values. Density is measured with the help of a digital vibrating-tube density meter (Anton Paar DSA 5000 M) as shown in Figure 1, with an accuracy of ± 0.02 K in temperature. It is calibrated with water at a particular temperature before the actual measurement. It measures the density of the pure components and their binary mixtures as well. The tube of the density meter was washed with water and then with acetone after every measurement.

Figure 1. Density meter DSA 5000 M

Figure 2. Modified Ubbelohde Viscometer

Figure 3. Digital water bath set up

Viscosity is measured with the help of a modified Ubbelohde viscometer, as shown in Figure 2, by using a digital stopwatch. Calibration of the viscometer is

done by using distilled benzene and cyclohexane at particular temperatures. The digital water bath, as shown in Figure 3, has been used to achieve different temperatures of the mixtures with an accuracy of ± 0.01 K. The sample was inserted in the viscometer and then dipped vertically in the water bath for about half an hour to achieve thermal equilibrium. Three readings of efflux time, i.e. time taken for the sample to pass through the viscometer, are taken, and the average value of the time is used for the calculation of viscosity. Equation 2 has been used for viscosity calculation is

$$v = \frac{\eta}{\rho} = At + \frac{B}{t} \qquad (2)$$

Where ν, **η**, ρ, and t represent kinematic viscosity, dynamic viscosity, density, and average time, respectively. A and B are constants that were calculated from the efflux times.

3.3. Correlations used

3.3.1. Density

Equation 3 has been used to calculate the excess molar volume (V^E) values of a binary mixture.

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho_m} - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right) \qquad (3)$$

where ρ_m represents the density of the binary mixture; x_1, ρ_1 and M_1 respectively represent the molar fraction, density and relative molecular mass of DMC; x_2, ρ_2 and M_2 represent the molar fraction, density, and relative molecular mass of *o*-xylene or *m*-xylene, respectively.

3.3.2. Viscosity

The experimentally determined values of viscosity were fitted to the McAllister model given by Eq. 4, where ν, ν_1, and ν_2 are the kinematic viscosity values of the mixture and pure component, respectively, x_1 and x_2 refer to the mole fractions, M_1, M_2 are the molecular weights of pure components, and A_{12}, A_{21} are the parameters.

$$\ln v = x_1^3 \ln \nu_1 + x_2^3 \ln \nu_2 + 3x_1^2x_2 \ln A_{12} + 3x_1x_2^2 \ln A_{21} - \ln\left[x_1 + x_2\frac{M_2}{M_1}\right] + 3x_1^2x_2 \ln\left[\frac{2 + \left(\frac{M_2}{M_1}\right)}{3}\right] + 3x_1x_2^2 \ln\left[\frac{1 + 2\left(\frac{M_2}{M_1}\right)}{3}\right] + x_2^3 \ln\left[\frac{M_2}{M_1}\right] \qquad (4)$$

Grunberg and Nissan Parameter, d, which is a measure of the strength of the interaction between components of the binary mixture, can be estimated using Eq. 5

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2 d \qquad (5)$$

Here, **η**, η_1 and η_2 represent the viscosity of the mixture, DMC and xylene, whereas x_1 and x_2 refer to the mole fractions of DMC and xylene. The deviation in viscosity was obtained by following Eq. 6

$$\Delta\eta = \eta_m - (x_1\eta_1 + x_2\eta_2) \qquad (6)$$

η_1 and η_2 are the viscosities of pure components 1 & 2, respectively, and η_m is the mixture viscosity

3.3.3. Excess Gibbs energy of activation

The excess Gibbs free energy of activation of flow can be calculated using Eq. 7.

$$\frac{G^E}{RT} = \ln(V_m\eta) - \sum_{i=1}^{2} x_i \ln(\eta_i V_i) \qquad (7)$$

Here, G^E is the excess Gibbs energy of activation, R is the universal gas constant, T is the temperature in Kelvin, V_1 and V_2 are the molar volumes of the pure components 1 and 2, and V_m is the molar volume of the mixture.

3.3.4. Redlich-Kister Equation

The excess molar volume, V^E, excess Gibbs energy of activation, ΔG^E, and deviations in viscosity, **Δη**, can be fitted to a Redlich-Kister type Eq. 8.

$$A = x_1x_2 \sum_{j=1}^{n} A_{(j-1)}(x_1 - x_2)^{j-1} \qquad (8)$$

Here, A is the property under consideration, $A_{(j-1)}$ is the polynomial coefficient, and n is the polynomial degree.

3.3.5. Standard deviation equation

The standard deviation in each case can be calculated using Eq. 9

$$\sigma(X) = \left[\frac{\sum(X_{exp} - X_{cal})}{(N - n)}\right] \qquad (9)$$

Here, X, N, and n represent the property under consideration, the number of data points, and the coefficients, respectively. Subscripts exp. and cal. means experimental and calculated.

4. Results and discussion

The results have been represented graphically in Figure 4-10. Table 2 and Table 3 contain the A_k coefficients of the Redlich-Kister polynomial equation and the Standard deviation used for fitting V^E, Δη and G^E in the case of a mixture of DMC (1) with o-xylene and m-xylene respectively.

Table 2. Coefficients (A_K) of the Redlich-Kister Equation and Standard Deviations (σ) for the fitting of (V^E), ($\Delta\eta$) and (G^E) for DMC(1) and *o*-xylene(2) at T= (298.15 – 313.15) K

T/K	A_0	A_1	A_2	A_3	σ
V^E					
298.15	0.0805	0.0472	-0.0108	-0.0310	0.0041
303.15	0.0974	0.0563	0.0073	-0.0218	0.0048
308.15	0.1375	0.1197	-0.0271	-0.0931	0.0040
313.15	0.1646	0.0921	-0.0414	-0.0409	0.0041
$\Delta\eta$					
298.15	0.0633	0.0117	0.0008	0.0001	0.0011
303.15	0.0592	0.0117	-0.0016	-0.0046	0.0010
308.15	0.0566	0.0112	-0.0002	-0.0022	0.0013
313.15	0.0515	0.0100	0.0016	-0.0029	0.0011
G^E					
298.15	0.4768	0.0999	0.0212	0.0095	0.0029
303.15	0.4824	0.1042	0.0077	-0.0161	0.0031
308.15	0.4916	0.0979	0.0310	0.026	0.0027
313.15	0.4918	0.1372	-0.0082	-0.1094	0.0023

Table 3. Coefficients (A_K) of the Redlich-Kister Equation and Standard Deviations (σ) for the fitting of (V^E), ($\Delta\eta$) and (G^E) for DMC(1) and *m*-xylene(2) at T= (298.15 – 313.15) K

T/K	A_0	A_1	A_2	A_3	σ
V^E					
298.15	0.0712	0.0676	0.0391	-0.0068	0.0026
303.15	0.0966	0.0705	0.0197	-0.0072	0.0026
308.15	0.1105	0.0638	0.0095	0.0133	0.0028
313.15	0.1241	0.0411	0.0031	0.0580	0.0031
$\Delta\eta$					
298.15	0.0034	0.0015	0.0007	-0.0019	0.0010
303.15	0.0030	0.0020	-0.0003	-0.0039	0.0013
308.15	0.0025	0.0008	-0.0004	-0.0011	0.0011
313.15	0.0019	0.0010	-0.0002	-0.0019	0.0012
G^E					
298.15	0.1896	0.0292	0.0055	-0.0094	0.0022
303.15	0.1918	0.0297	0.0049	-0.0110	0.0024
308.15	0.1938	0.0248	0.0046	0.0001	0.0021
313.15	0.1944	0.0274	0.0023	-0.0088	0.0017

From the experimental data, it was concluded that the density of both binary liquid mixtures, i.e. DMC + *o*-xylene and DMC + *m*-xylene, decreases with an increase in temperature and increases as the mole fraction of DMC increases. The highest value of density of the mixture has been observed at the highest mole fraction of DMC (i.e. x= 0.9649) and the lowest temperature (298.15 K) in both cases. The density has been decreased by 1.84% in case of both mixtures with the highest mole fraction of DMC when the temperature has been increased from 298.15 to 313.15K.

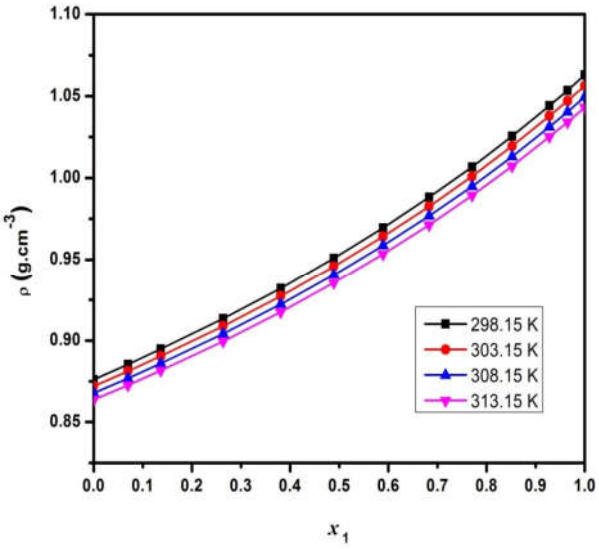


Figure 4: Density, ρ vs. mole fraction, x_1 of DMC(1) + *o*-xylene(2) at 298.15 K, 303.15 K, 308.15 K and 313.15 K.

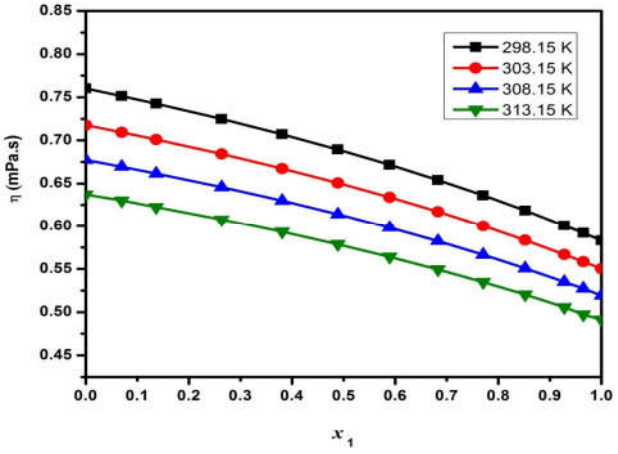


Figure 5: Viscosity, η vs. mole fraction, x_1 of DMC (1) + *o*-xylene (2) at 298.15 K,303.15 K, 308.15 K and 313.15 K.

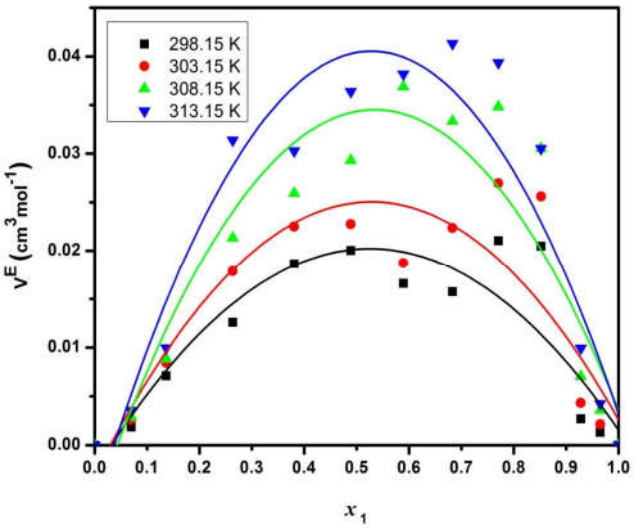


Figure 6: Excess Molar Volume, V^E vs. mole fraction, x_1 of DMC (1) + *o*-xylene (2) at298.15 K, 303.15 K, 308.15 K and 313.15 K.

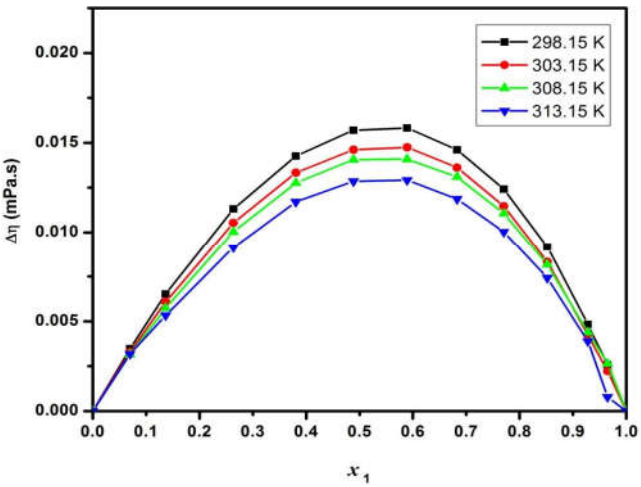


Figure 7: Deviation in Viscosity, Δη vs. mole fraction, x_1 of DMC(1) + *o*-xylene(2) at 298.15 K, 303.15 K, 308.15 K and 313.15 K

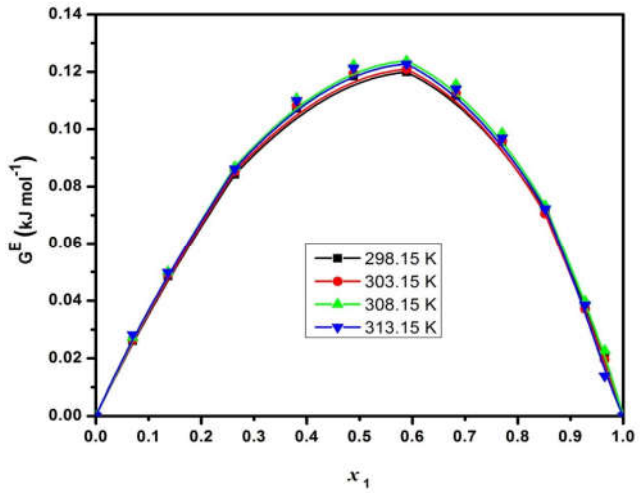


Figure 8: Excess Gibbs Energy of Activation, G^E vs. mole fraction, x_1 of DMC(1) + *o*-xylene(2) at 298.15 K, 303.15 K, 308.15 K and 313.15 K.

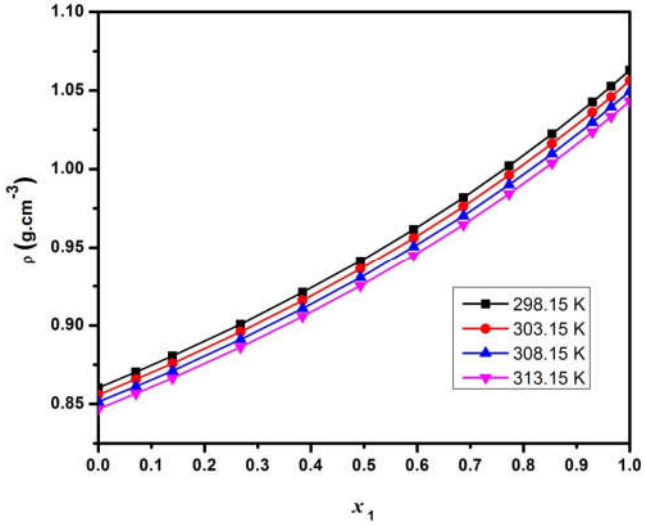


Figure 9: Density, ρ vs. mole fraction, x_1 of DMC (1) + *m*-xylene (2) at 298.15 K,303.15 K, 308.15 K and 313.15K

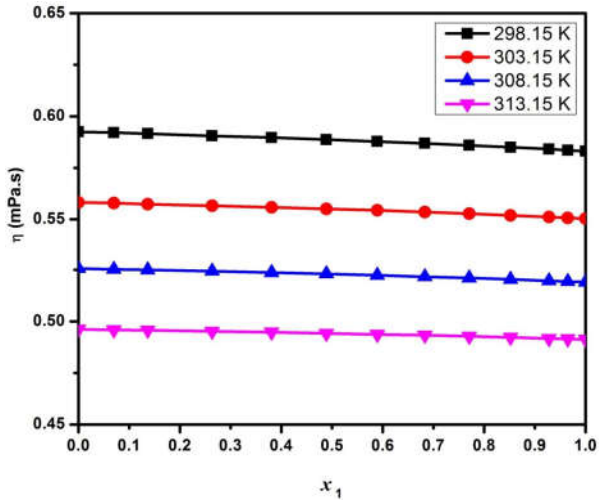


Figure 10: Viscosity, η vs. mole fraction, x_1 of DMC(1) + *m*-xylene(2) at 298.15 K,303.15 K, 308.15 K and 313.15 K

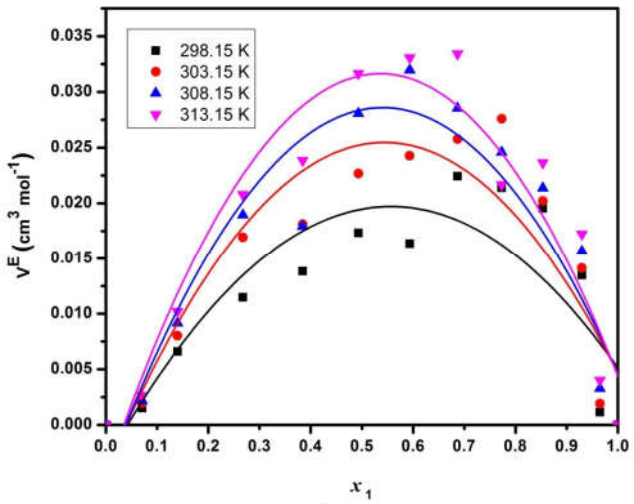


Figure 11: Excess Molar Volume, V^E vs. mole fraction, x_1 of DMC(1) + *m*-xylene(2) at298.15 K, 303.15 K, 308.15 K and 313.15 K

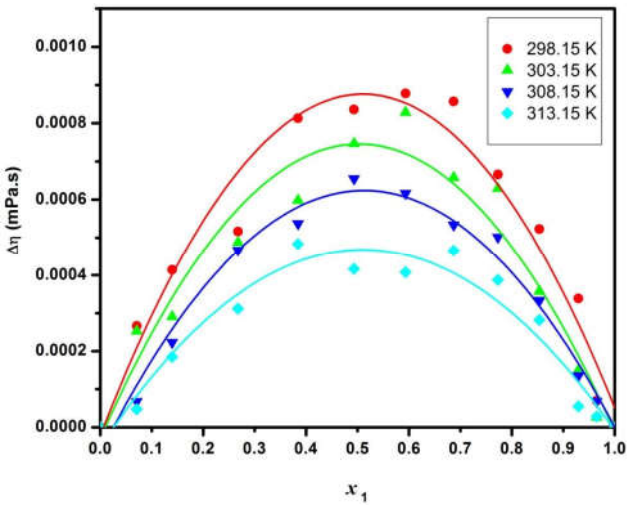


Figure 12: Deviation in Viscosity, $\Delta\eta$ vs. mole fraction, x_1 of DMC(1) + *m*-xylene(2) at 298.15 K, 303.15 K, 308.15 K and 313.15 K

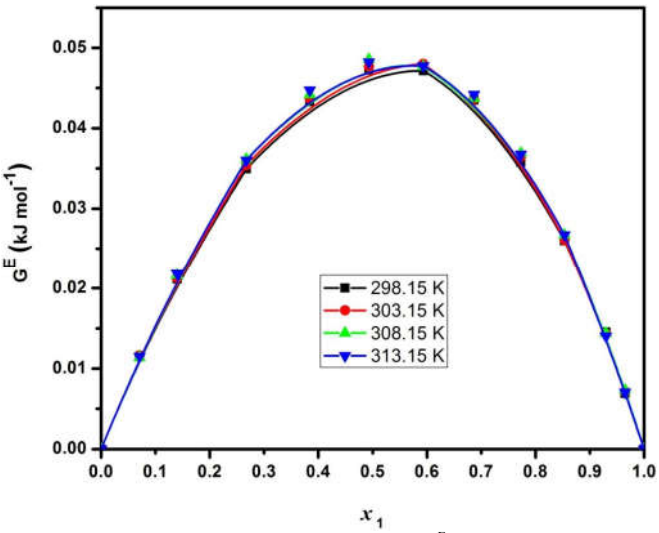


Figure 13: Excess Gibbs Energy of Activation, G^E vs. mole fraction, x_1 of DMC(1) + *m*-xylene (2) at 298.15 K, 303.15 K, 308.15 K & 313.15 K

Figures 6 and 10 graphically represent the excess molar volume data for the binary liquid mixtures of DMC + *o*-xylene and *m*-xylene, respectively. Both figures show the positive values of $V^{E,}$ which indicate that there is expansion in volume on mixing. At the same time, it can be seen that V^E curves are shifted regularly with

increasing temperature (i.e. V^E becomes more positive at higher temperatures). The binary liquid mixture of *o*-xylene shows more positive deviations than *m*-xylene.

It has been suggested that V^E is the result of the contributions from several opposing effects, which may be divided into three types: physical, chemical, and structural effects. Physical effects make a positive contribution to V^E, whereas chemical and structural effects make a negative contribution. The V^E values are all positive for these binary mixtures. It indicates that the physical contributions are dominant.

From the experimental data, it has been observed that the viscosity of both binary liquid mixtures decreases with an increase in temperature and an increase in the mole fraction of DMC. Decrements of 15.9 and 15.7 % have been observed in the case of DMC + *o*-xylene and DMC + *m*-xylene mixtures, respectively, with the highest mole fraction (x=0.9649) when the temperature has been increased from 298.15K to 313.15 K.

With an increase in the mole fraction of DMC from 0.0698 to 0.9649, viscosity is decreased by 21% at a maximum temperature of 313.15K in the DMC+o-xylene mixture. Whereas, a negligible decrease of 0.88 % has been observed when the mole fraction of DMC changed from 0.0709 to 0.9649 at a maximum temperature of 313.15K in the case of a DMC+ m-xylene mixture.

Figures 7 and 12 suggest that deviation in viscosity, $\Delta\eta$ values are slightly positive for both the binary mixtures of DMC + *o*-xylene and DMC + *m*-xylene at all temperatures, which indicates very little deviation from ideality.

Figures 8 and 13 show that G^E values are positive over the entire composition range for both the binary mixtures at selected temperatures. With the increase in temperature from 298.15 K to 313.15 K, G^E values increase but not significantly. The value of G^E increases more in the case of DMC + *o*-xylene as compared to DMC + *m*-xylene.

5. Conclusion

The prepared binary liquid mixtures of DMC with *o*-xylene and *m*-xylene have been found stable during experimentation. The Density ρ and viscosity η have been measured experimentally over the whole composition range at four different temperatures (298.15 K, 303.15 K, 308.15 K & 313.15 K) and atmospheric pressure. By using these values, Excess molar volume, V^E, deviations in viscosity, $\Delta\eta$ and excess Gibbs free energy of activation, G^E have been calculated. The V^E, $\Delta\eta$, and G^E have been fitted to the Redlich-Kister polynomial equation, and the A_k coefficients, as well as standard deviation values, have been estimated. From the experimental data, it was concluded that the density of both binary liquid mixtures, i.e. DMC + *o*-xylene and DMC + *m*-xylene, decreases with an increase in temperature and increases as the mole fraction of DMC increases. From the experimental data, it has been observed that the viscosity of both binary liquid mixtures decreases with an increase in temperature and an increase in the mole fraction of DMC.

Acknowledgements

Authors are thankful to the Chairperson, Dr SSB UICET, PU, Chandigarh, for providing the necessary set-up and facilities to carry out experimental work.

Nomenclature

DMC	:	Dimethyl Carbonate
V^E	:	Excess Molar Volume
ρ	:	Density
η	:	Viscosity
$\Delta\eta$	:	Deviation in Viscosity
G^E	:	Excess Gibbs energy of activation
R	:	Universal Gas Constant
T	:	Absolute temperature
σ	:	Standard deviation
A_0, A_1, A_2, A_3	:	Parameters of the Redlich-Kister Equation

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