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TRANSPORT PROPERTIES OF BINARY LIQUID MIXTURES IN MEK WITH BROMOBENZENE

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ABSTRACT

Viscosities and densities for MEK with Bromobenzene have been measured as a function of mole fraction at atmospheric pressure and at temperatures of 303.15 K, 308.15 K, and 313.15 K. The calculated deviations in viscosities and excess volumes were reported. McAllister's three-body-interaction model, Krishnan and Laddha model and the Jouyban Acree model were used to correlate the kinematic viscosity of the systems. The excess volume data was fitted by means of the Redlich-Kister equation. It was found that in all cases the experimental data obtained fitted with the values correlated by the corresponding model very well.

Keywords: *Bromobenzene, Density, Excess molar volume, Viscosity and Viscosity deviation.*

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1. INTRODUCTION

The physical properties and the thermodynamic behaviour of binary mixtures have been studied for many reasons, one of the most important of which is that these properties may provide information about molecular interactions [1-6]. This paper is part of our systematic program of research on the measurement of physical and transport properties of binary liquid mixtures containing MEK with Bromobenzene, which are widely used in the extraction of aromatics in petrochemical processing. In this work, we present density and viscosity data for the binary systems MEK with Bromobenzene at 303.15, 308.15 and 313.15K and over the whole mole fraction range.

Literature survey showed that no measurements have been previously reported for the MEK with Bromobenzene binary mixture. The objective of the present investigation was to find out the density (ρ), viscosity (η), Excess molar volume (VE) and viscosity deviation (Δη) of pure MEK and Bromobenzene as well as for the binary system constituted by these chemicals at 303.15, 308.15 and 313.15K.

The experimental values were used to calculate excess molar volumes and the deviation in viscosity over the entire mole fraction range for the binary mixtures. The computed quantities have been fitted to the "McAllister, (1960)" model[7], "Krishnan and Laddha"model[8], "Jouyban Acree" model[10] and Redlich-kister

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model[9].The deviation values have been fitted to "Redlich-Kister,(1948)" equation.The results are discussed in terms of the molecular interactions[11-14].

2. MATERIALS AND METHODS

2.1 Materials

MEK and Bromobenzene were purified by fractional distillation and drying. For purity check, the densities and viscosities of the pure liquids were measured at 25 ± 0.010K. The mean of several repeat measurements compared with the corresponding literature value within allowable limits (Table 1). Redistilled and deionized water which showed an electrical conductivity <7.0x10-7 mhos cm-1 was used for checking the instruments and calibrating the pycnometer for density measurements. Care was taken to expel all the dissolved air from the water used before calibration.

2.2 Apparatus and Procedure

The densities of the pure components and their mixtures were measured with a high-precision Ostwold-Sprengal- type pycnometer[15-20] having a bulk volume of 25 cm^3 and an internal diameter of the capillary of about 1 mm whose measurement cell temperature was controlled automatically to within ± 0.010 K of the selected value. Before each series of measurements, the instrument was calibrated at atmospheric pressure with double-distilled water and dry air. Densities both in water and dry air at the various working temperatures were given by the manufacturer in the instruction manual. The calibration was accepted if the measurements were within 5×10^{-5} $g.cm^{-3}$ of the published values. The uncertainty in the density measurements was 5×10^{-5} $g.cm^{-3}$. Density measurements were reproducible to 3×10^{-5} $g.cm^{-3}$. The liquid mixtures were prepared by weight using a BP210s balance that was accurate to within (0.01 mg). The average uncertainty in the mole fraction of the mixtures was estimated to be less than 0.0001. The molar excess volumes were calculated from composition-density data with an uncertainty better than 0.002 $cm^3.mol^{-1}$. All molar quantities were based on the IUPAC relative atomic mass table.

The viscosities of the pure liquids and the mixtures were measured at atmospheric pressure and at different temperatures using Oswald Viscometer[21-26] supplied by SAI Scientific Company, Madras. The viscometer was immersed in a well-stirred water bath (Lauda, Germany) with temperature control to within 0.01 K. An electronic digital stopwatch with a readability of 0.01s was used for flow time measurements. Experiments were repeated a minimum of five times at each temperature for all compositions, and the results were averaged. The viscosity η of the liquid was then calculated from the following relationship

$$\nu = (at) - (b/t) \qquad (1)$$

Where ν is the kinematic viscosity, a and b is the constants and t is the time.
The calibration of the viscometer was carried out with double-distilled water and double-distilled benzene. Care was taken to reduce evaporation during the measurements. The uncertainty in the values is within 0.003 Pa.s.
In the experiment, the density and viscosity for the one composition sample were measured at different temperatures. Densities and viscosities of pure compounds are reported in Table 1 together with the corresponding literature values.

3. RESULTS AND DISCUSSION

3.1 Viscosity deviation

Viscosity deviations [27-33] were calculated from our measurements according to the following equation

$$\Delta\eta=\eta_{12}-(x_1\eta_1 + x_2\eta_2) \quad (2)$$

Where η, η_1, and η_2 are the dynamic viscosities of the mixture and those of the pure components 1 and 2, respectively.

The kinematic viscosities were correlated by means of the McAllister model [34-35] considering a three-body-interaction model, which for two-component mixtures gives

$$\ln\nu = x_1^3\ln\nu_1 + 3x_1^2x_2\ln\nu_{12} + 3x_1x_2^2\ln\nu_{21} + x_2^3\ln\nu_2 - \ln(x_1 + x_2M_2/M_1) + 3x_1^2x_2\ln((2+M_2/M_1)/3)+x_2^3\ln(M_2/M_1)+3x_1x_2^2 2\ln((1+2M_2/M_1)/3) \quad (3)$$

Where ν refers to the kinematic viscosity of the mixture of components 1 and 2 having mole fractions x_1 and x_2 respectively. $\boldsymbol{\nu_1}$ and $\boldsymbol{\nu_2}$ refers to the kinematic viscosity of pure liquids 1 and 2 respectively. $\boldsymbol{\nu_{12}}$ and $\boldsymbol{\nu_{21}}$ represent the interaction parameters obtained by multiple regression analysis. M_1 and M_2 refer to the molecular weight of the two components respectively. The values of the parameters $\boldsymbol{\nu_{12}}$ and $\boldsymbol{\nu_{21}}$are given in Table 4.

Krishnan and Laddha have proposed an equation to predict viscosities of binary liquid mixtures based on Eyring's theory of absolute reaction rate. The equation is as follows

$$\ln\upsilon_{mix}= x_1\ln\upsilon_1+x_2\ln\upsilon_2 - 2.303x_1x_2\,(A+B(x_1-x_2))\ln(x_{1M1}+x_2M_2)+x_1\ln M_1+x_2\ln M_1 \quad (4)$$

The values of parameters A and B are given in table 5.

Jouyban [36] proposed a model for correlating the viscosity of binary liquid mixture at various temperatures.

$$\ln\upsilon_{mix}=x_1\ln\upsilon_1+x_2\ln\upsilon_2+(x_1x_2/T)\sum a_i(x_1-x_2)^i \quad (5)$$

The values of parameter a_i are given in table 6.

The percentage deviation was calculated by

$$d = ((\nu_{exp} - \nu_{cal})/\nu_{cal})*100 \quad (6)$$

The average deviation (AD) was calculated from the relationship

$$AD = (\sum d^2/N)^{1/2} \quad (7)$$

In figure 1, the shape of deviation in viscosity observed in the MEK with Bromobenzene systems are attributed to varying interaction between a relatively large negative contribution due to chemical and structural effect as reported by Nikam [37-40].The values of Δη for the system containing MEK with Bromobenzene are asymmetrical and are negative throughout the whole concentration range at all temperatures. The viscosity of a mixture strongly depends on the entropy of the mixture, which is related with liquid structure and enthalpy and consequently with molecular interactions between the components of the mixture. Therefore, the viscosity deviation depends on molecular interactions as well as on the size and shape of the molecules.

3.2 Excess molar volume

The excess molar volumes [41-43] (V^E) can be computed from experimental density data using the relationship

$$V^E = (x_1M_1 + x_2M_2)/\rho_m - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (8)$$

Where x_1 and x_2 refers to the mole fraction of components1 and 2. ρ_1 and ρ_2 refer to the density of components 1 and 2. ρ_m is the density of the mixture. For each mixture the excess molar volumes were fitted with Redlich-Kister equation

$$V^E = x_1x_2 \sum a_{i-1}(x1 - x2)^{i-1} \quad (9)$$

The coefficients a_{i-1} and standard deviations are listed in Table 7 and 8.The standard deviation was calculated by the following equation

$$\sigma(V^E) = [\sum(V^E_{exp} - V_{cal}{}^E)^2/(N - m)]^{1/2} \quad (10)$$

Where N is the number of experimental points and m is the number of coefficients in the corresponding equations. In figure 2, The V^E values are positive except at higher mole fractions of Toluene. Positive values are attributed to changes in a free volume in the mixture of electron donor-acceptor-type interactions between MEK with Bromobenzene. Excess molar volumes of binary mixtures are positive and increase slightly with increasing temperature. When aromatics, which exist in a highly associated form in the pure state. The mono merization occurs and new specific interactions appear in the solution. The disruption of the hydrogen-bonded aromatic structure gives rise to a positive contribution to V^E, which depends: (i) on the dielectric constant of the MEK of chain length15 and (ii) on the dielectric constant of the Bromobenzene of chain length 16 and degree of branching in the aromatic which decrease the self-association in the pure state. On the other hand, the interactions between unlike molecules in both systems are surely weaker than the sum of the interactions between like molecules. These effects produce positive

excess molar volumes. Excess molar volumes for each binary mixture increase with increasing temperature.

4. CONCLUSION

The McAllister model, Krishnan and Laddha model and Jouyban Acree model are considered one of the best for correlating viscosity with composition. The models were used to correlate the kinematic viscosity–composition data for the binary systems, MEK with Bromobenzene, The kinematic viscosity correlated by McAllister, Krishnan - Laddha and Jouyban Acree model were in excellent agreement with the experimental data. The Redlich - Kister equation is considered one of the best for correlating excess volume with composition. The excess volumes calculated by RedlichKister equation were in excellent agreement with experimental data.

Densities and viscosities for MEK with Bromobenzene at temperatures of (303.15, 308.15 and 313.15) K, have been experimentally determined over the entire mole fraction range. The excess molar volumes were correlated using the Redlich- Kister polynomial equation. The excess molar volumes for the binary mixtures of MEK with Bromobenzene are positive over the whole composition range. On the contrary, the deviations in viscosity for these systems at selected temperatures are all negative over the entire composition. The absolute viscosity deviations for MEK with Bromobenzene are much larger.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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Tab.1. Comparison of Experimental and Literature Values of Densities (ρ) and Viscosities (η) for pure compounds.

Liquid	Temperature T/K	ρ		η	
		Exptl	Lit	Exptl	Lit
	303.15	0.7948	0.7949	0.3559	0.3558
MEK	308.15	0.8656	0.8658	0.6150	0.6149
	313.15	0.8670	0.8669	0.6069	0.6068
	303.15	1.4888	1.4889	0.6242	0.6246
Bromobenzene	308.15	0.7894	0.7889	0.3462	0.3474
	313.15	0.7946	0.7948	0.3369	0.3401

Tab.2. Experimental Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations (Δη) for MEK with Bromobenzene at Different Temperatures.

X_1	ρ	V^E	γ	η	Δη
		T=303.15K			
0	1.4889	0.0000	0.9850	1.4665	0.0000
0.1143	1.2631	0.0143	0.9815	1.2397	-0.0998
0.2251	1.0158	0.0223	0.9361	0.9508	-0.1953
0.3325	0.9931	0.0264	0.9010	0.8947	-0.2589
0.4365	0.9542	0.0288	0.8952	0.8542	-0.2699
0.5375	0.9268	0.0299	0.8751	0.8110	-0.2538
0.6355	0.9026	0.0298	0.8302	0.7493	-0.2256
0.7306	0.9005	0.0287	0.8083	0.7278	-0.1968
0.8229	0.8504	0.0251	0.7759	0.6598	-0.1559
0.9127	0.8209	0.0183	0.7292	0.5986	-0.1110
1	0.7948	0.0000	0.3559	0.2828	0.0000
		T=308.15K			
0	1.4680	0.0000	0.9530	1.3990	0.0000
0.1143	1.2309	0.0095	0.8927	1.0988	-0.0599
0.2251	1.0086	0.0157	0.7765	0.7831	-0.1268
0.3325	0.9917	0.0209	0.6431	0.6377	-0.1959
0.4365	0.8951	0.0243	0.6298	0.5637	-0.2408
0.5375	0.8745	0.0264	0.5994	0.5241	-0.2299
0.6355	0.8291	0.0260	0.5568	0.4616	-0.1989
0.7306	0.8028	0.0245	0.4811	0.3862	-0.1686
0.8229	0.7998	0.0215	0.4423	0.3537	-0.1299
0.9127	0.7985	0.0129	0.3978	0.3176	-0.0865
1	0.7890	0.0000	0.3460	0.2729	0.0000
		T=313.15K			
0	1.4748	0.0000	0.8744	1.2895	0.0000
0.1143	1.1408	0.0067	0.8294	0.9461	-0.0367
0.2251	1.0002	0.0125	0.7856	0.7857	-0.0932
0.3325	0.9657	0.0169	0.6183	0.5970	-0.1586
0.4365	0.8543	0.0197	0.5899	0.5039	-0.1998
0.5375	0.8221	0.0216	0.5505	0.4525	-0.1876
0.6355	0.8019	0.0223	0.5289	0.4241	-0.1598
0.7306	0.7999	0.0205	0.5093	0.4073	-0.1283
0.8229	0.7890	0.0175	0.4863	0.3836	-0.0958
0.9127	0.7877	0.0101	0.4301	0.3387	-0.0599
1	0.7844	0.0000	0.3368	0.2641	0.0000

Tab.3. Predicted kinematic viscosities and Excess molar volume For MEK with Bromobenzene at Different Temperatures.

X_1	γ_{expt}	γ_{pred} (McAllister Model)	γ_{pred} (Jouyban- Acree Model)	γ_{pred} (K-L model)	V^E (pred) (R-K model) (cc/gmole)
		T=303.15^{0}K			
0	0.9850	0.9850	0.9850	0.9850	0
0.1143	0.9815	0.9813	0.9815	0.9810	0.0143
0.2251	0.9361	0.9362	0.9360	0.9359	0.0226
0.3325	0.9010	0.9011	0.9015	0.9010	0.0265
0.4365	0.8952	0.8953	0.8950	0.8952	0.0288
0.5375	0.8751	0.8750	0.8752	0.8756	0.0298
0.6355	0.8302	0.8302	0.8312	0.8310	0.0296
0.7306	0.8083	0.8083	0.8083	0.8083	0.0286
0.8229	0.7759	0.7758	0.7759	0.7756	0.0250
0.9127	0.7292	0.7293	0.7296	0.7292	0.0183
1	0.3559	0.3559	0.3559	0.3559	0
		T=308.15^{0}K			
0	0.8744	0.8744	0.8744	0.8744	0
0.1143	0.8294	0.8291	0.8292	0.8290	0.0067
0.2251	0.7856	0.7855	0.7864	0.7865	0.0123
0.3325	0.6183	0.6179	0.6180	0.6183	0.0170
0.4365	0.5899	0.5898	0.5896	0.5888	0.0197
0.5375	0.5505	0.5515	0.55502	0.5525	0.0213
0.6355	0.5289	0.5279	0.5086	0.5298	0.0223
0.7306	0.5093	0.5094	0.5091	0.5093	0.0204
0.8229	0.4863	0.4865	0.4860	0.4863	0.0173
0.9127	0.4301	0.4301	0.4299	0.4312	0.0102
1	0.3368	0.3368	0.3368	0.3368	0.0000
		T=313.15^{0}K			
0	0.8744	0.8744	0.8744	0.8744	0
0.1143	0.8294	0.8291	0.8292	0.8290	0.0067
0.2251	0.7856	0.7855	0.7864	0.7865	0.0123
0.3325	0.6183	0.6179	0.6180	0.6183	0.0170
0.4365	0.5899	0.5898	0.5896	0.5888	0.0197
0.5375	0.5505	0.5515	0.55502	0.5525	0.0213
0.6355	0.5289	0.5279	0.5086	0.5298	0.0223
0.7306	0.5093	0.5094	0.5091	0.5093	0.0204
0.8229	0.4863	0.4865	0.4860	0.4863	0.0173
0.9127	0.4301	0.4301	0.4299	0.4312	0.0102
1	0.3368	0.3368	0.3368	0.3368	0.0000

Tab.4. Parameters of McAllister constants for MEK with Bromobenzene System at 303.15K, 308.15K and 313.15K.

Temperature T/K	ν_{12}	ν_{21}	SD
303.15	0.9986	0.8551	0.0226
308.15	0.9531	0.7983	0.3605
313.15	0.8754	0.7796	0.0516

Tab.5. Parameters of the Krishnan and Laddha Constants and standard deviations S for the viscosity of MEK with Bromobenzene.

Temperature T/K	A_1	A_2	A_3	SD
303.15	-0.4166	0.0902	0.0680	0.7164
308.15	0.0647	0.0920	-0.0416	0.2236
313.15	-0.7234	0.0104	-0.0047	0.1857

Tab.6. Parameters of the Jouyban Acree model Constants and standard deviations S for the viscosity of MEK with Bromobenzene.

Temperature T/K	A_1	A_2	A_3	A_4	SD
303.15	63.0354	-78.3152	0.5137	68.98	0.7776
308.15	-74.6520	-4.6248	188.5796	-17.9301	0.54772
313.15	151.8479	8.9628	-76.2514	-36.5813	0.06879

Tab.7. Parameters of the Redlich Kister Constants and standard deviations S for Excess Volume of MEK with Bromobenzene.

Temperature T/K	A_1	A_2	A_3	A_4	A_5	SD
303.15	0.8640	0.7301	-0.7625	0.7023	-0.9075	0.9319
308.15	-0.4169	-0.2505	0.5614	0.2918	-0.3687	0.6138
313.15	-0.4078	0.2014	0.4325	0.7968	-0.3529	0.6333

Tab.8. Parameters of the Redlich - Kister Constants and standard deviations S for Viscosity Deviation of MEK with Bromobenzene

Temperature T/K	A_1	A_2	A_3	A_4	A_5	SD
303.15	-0.7916	0.8916	-0.7423	-0.6961	0.7701	1.2655
308.15	-0.7571	0.9738	-0.5387	-0.6378	0.6811	1.1738
313.15	-0.7416	0.8601	-0.4801	-0.5011	0.5976	1.1405

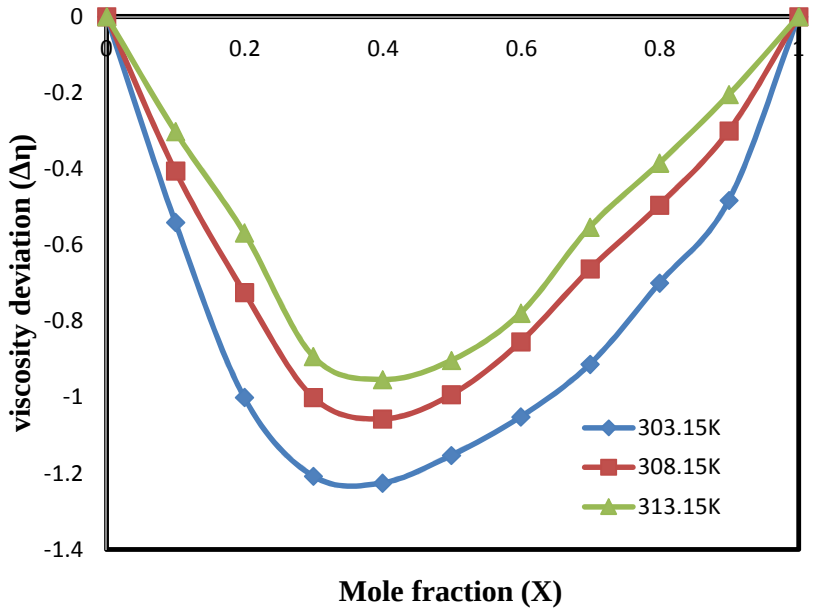

Fig.1. Viscosity deviation with mole fraction for the MEK with Bromobenzene at 303.15K, 308.15K and 313.15K.

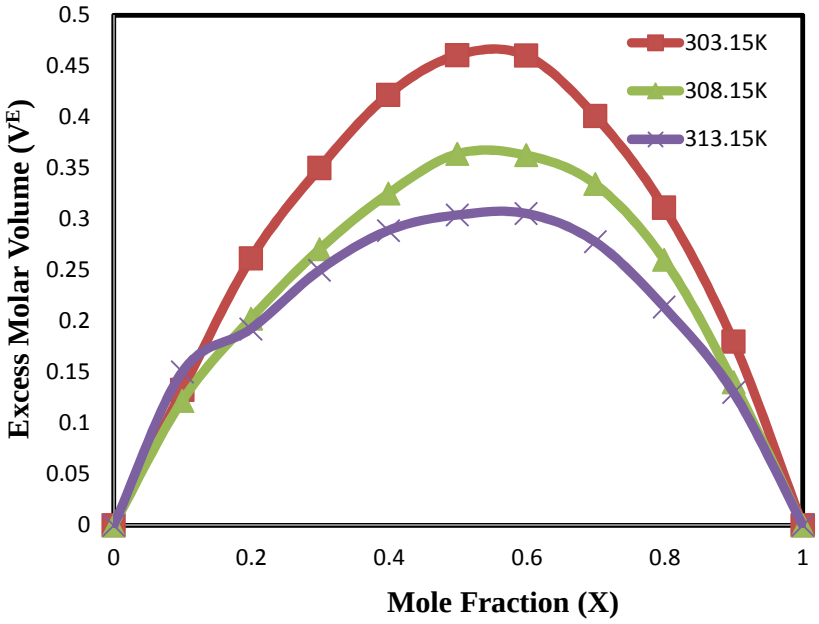

Fig.2. Excess molar volume with mole fraction for the system MEK with Bromorobenzene at 303.15K, 308.15K and 313.15K.

SYMBOLS

ν	[cS]	Kinematic Viscosity
H	[kg/m.sec]	Dynamic Viscosity
Δη	[cP]	Viscosity Deviation
V^E	[cm^3/gmol]	Excess molar volume
P	[g/cc]	Density
ν_{12}	-	Interaction Parameter
ν_{21}	-	Interaction Parameter
X_1		Mole fraction of component 1
X_2	[kg/kg.mole]	Mole fraction of component 2
M_1	[kg/kg.mole]	Molecular weight of component 1
M_2		Molecular weight of component 2
Σ	-	Standard deviation
N	-	Number of data experimental points
M	-	Number of coefficients
ρ_m	[g/cc]	Density of mixture
ρ_1	[g/cc]	Density of component 1
ρ_2	[g/cc]	Density of component 2
a_{i-1}	-	Redlich-kister coefficient
AD	-	Average deviation
d	-	Percentage deviation
MEK	-	Methyl Ethyl Ketone

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