



**THERMODYNAMIC PROPERTIES OF BINARY LIQUID
MIXTURE OF TOLUENE WITH BENZENE**

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ABSTRACT

The experimental densities and viscosities of Toluene with benzene at temperatures of 303.15, 308.15 and 313.15K were determined using a vibrating-tube density meter and Oswald Viscometer. From these data, excess molar volumes and viscosity deviations have been calculated. The computed quantities have been fitted to the respective models to derive the coefficients and estimate the standard error values. The results are discussed in terms of the molecular interactions.

KEYWORDS: Benzene, Density, Excess molar volume, viscosity, viscosity deviation and Toluene.



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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 interactions¹⁻⁶. This paper is part of our systematic program of research on the measurement of physical and transport properties of binary liquid mixtures containing Toluene with benzene, 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 Toluene with benzene 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 Toluene with Benzene binary mixture. The objective of the present investigation was to find out the density (ρ), viscosity (η), Excess molar volume (V^E) and viscosity deviation ($\Delta\eta$) of pure Toluene with Benzene 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)" model⁷, "Krishnan and Laddha" model⁸, "Jouyban Acree" model¹⁰ and Redlich-kister model⁹. The deviation values have been fitted to "Redlich-Kister,(1948)" equation. The results are discussed in terms of the molecular interactions¹¹⁻¹⁴.

MATERIALS AND METHODS

Materials

Toluene and benzene were purified by fractional distillation and drying. For purity check, the densities and viscosities of the pure liquids were measured at $25 \pm 0.01^\circ\text{K}$. 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 $<7.0 \times 10^{-7}$ 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.

Apparatus and Procedure

The densities of the pure components and their mixtures were measured with a high-precision Ostwald-Sprengal-type pycnometer¹⁵⁻²⁰ 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 $\pm 0.01^\circ\text{K}$ 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 $5 \times 10^{-5} \text{ g.cm}^{-3}$ of the published values. The uncertainty in the density measurements was $5 \times 10^{-5} \text{ g.cm}^{-3}$. Density measurements were reproducible to $3 \times 10^{-5} \text{ g.cm}^{-3}$. 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 $0.002 \text{ cm}^3.\text{mol}^{-1}$. 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 Viscometer²¹⁻²⁶ 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

$$v = (at) - (b/t) \quad (1)$$

Where v 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.

RESULTS AND DISCUSSION

Viscosity deviation

Viscosity deviations²⁷⁻³³ 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 model³⁴⁻³⁵ considering a three-body-interaction model, which for two-component mixtures gives

$$\ln v = x_1^3 \ln v_1 + 3x_1^2 x_2 \ln v_{12} + 3x_1 x_2^2 \ln v_{21} + x_2^3 \ln v_2 - \ln(x_1 + x_2 M_2 / M_1) + 3x_1^2 x_2 \ln((2+M_2/M_1)/3) + x_2^3 \ln(M_2/M_1) + 3x_1 x_2^2 \ln((1+2M_2/M_1)/3) \quad (3)$$

Where v refers to the kinematic viscosity of the mixture of components 1 and 2 having mole fractions x_1 and x_2 respectively. v_1 and v_2 refers to the kinematic viscosity of pure liquids 1 and 2 respectively. Where v_{12} and v_{21} are the interaction parameters calculated by multiple regression analysis. M_1 and M_2 are the molecular weight of Toluene and Benzene. The values of the v_{12} and v_{21} are represent 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

$$\ln u_{\text{mix}} = x_1 \ln u_1 + x_2 \ln u_2 - 2.303 x_1 x_2 (A + B(x_1 - x_2)) \ln(x_1 M_1 + x_2 M_2) + x_1 \ln M_1 + x_2 \ln M_2 \quad (4)$$

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

Jouyban³⁶ proposed a model for correlating the viscosity of binary liquid mixture at various temperatures.

$$\ln u_{\text{mix}} = x_1 \ln u_1 + x_2 \ln u_2 + (x_1 x_2 / T) \sum a_i (x_1 - x_2)^i \quad (5)$$

The values of parameter a_i are given in table 6. The percentage deviation was calculated by

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

The average deviation (AD) was calculated from the relationship

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

In Graph 1, the shape of deviation in viscosity observed in the Toluene with benzene systems are attributed to varying interaction between a relatively large negative contribution due to chemical and structural effect as reported by Nikam³⁷⁻⁴⁰. The values of $\Delta\eta$ for the system containing Toluene with benzene 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.

Excess molar volume

The excess molar volumes⁴¹⁻⁴³ (V^E) can be computed from experimental density data using the relationship

$$V^E = (x_1 M_1 + x_2 M_2) / \rho_m - (x_1 M_1 / \rho_1 + x_2 M_2 / \rho_2) \quad (8)$$

Where x_1 and x_2 refers to the mole fraction of components 1 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

$$V^E = x_1 x_2 \sum a_{i-1} (x_1 - x_2)^{i-1} \quad (9)$$

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

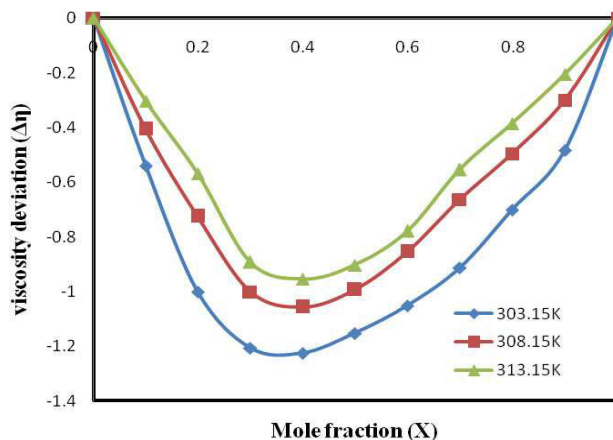
$$\sigma(V^E) = [\sum (V_{\text{exp}}^E - V_{\text{cal}}^E)^2 / (N - m)]^{1/2} \quad (10)$$

Where N is the number of experimental points and m is the number of coefficients in the corresponding equations. In Graph 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 Toluene with Benzene. 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 toluene of chain length 15 and (ii) on the dielectric constant of the benzene 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.

Graph 1
Viscosity deviation with mole fraction for the toluene with benzene at 303.15K, 308.15K and 313.15K



Graph 2
Excess molar volume with mole fraction for the system Toluene with Benzene at 303.15K, 308.15K and 313.15K

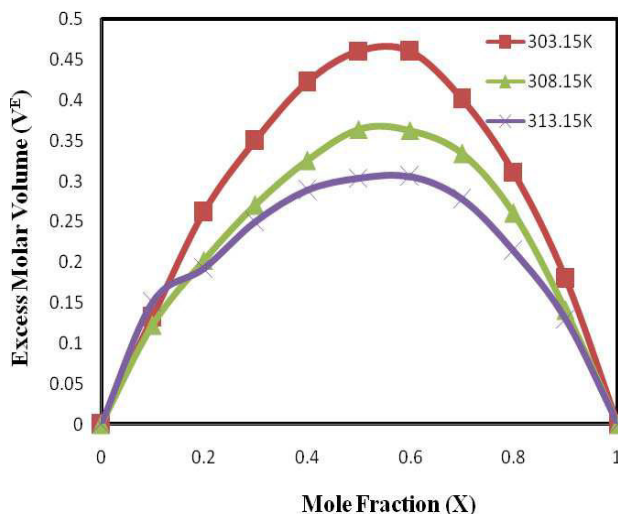


Table 1
Comparison of Experimental and Literature Values of Densities (ρ) and Viscosities (η) for pure compounds.

Liquid	Temperature T/K	P		η	
		Exptl	Lit	Exptl	Lit
Toluene	303.15	0.3559	0.3558	0.7946	0.7948
	308.15	0.8656	0.8658	0.6150	0.6149
	313.15	0.8670	0.8669	0.6069	0.6068
Benzene	303.15	0.8671	0.8670	0.6242	0.6246
	308.15	0.7894	0.7889	0.3462	0.3474
	313.15	0.7946	0.7948	0.3369	0.3401

Table 2
Experimental Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations ($\Delta\eta$) for Toluene with Benzene at Different Temperatures

X_1	ρ	V^E	γ	η	$\Delta\eta$
T=303.15K					
0.0000	0.8671	0.0000	0.6242	0.5412	0.0000
0.0993	0.8508	0.1325	0.6096	0.5186	-0.5402
0.1989	0.8460	0.2615	0.6057	0.5124	-1.0004
0.2985	0.8208	0.3501	0.5946	0.4305	-1.2068
0.3983	0.8017	0.4220	0.5185	0.4156	-1.2249
0.4982	0.8007	0.4609	0.5095	0.4079	-1.1523
0.5983	0.8004	0.4606	0.4947	0.3959	-1.0512
0.6985	0.7994	0.4014	0.4572	0.3345	-0.9132
0.7988	0.7971	0.3113	0.4197	0.3144	-0.7003
0.8993	0.7968	0.1802	0.3946	0.3099	-0.4829
1.0000	0.7949	0.0000	0.3559	0.2829	0.0000
T=308.15K					
0.0000	0.8656	0.0000	0.6150	0.5323	0.0000
0.0993	0.8489	0.1220	0.6049	0.5134	-0.4053
0.1989	0.8216	0.2025	0.6012	0.4939	-0.7243
0.2985	0.8172	0.2705	0.6001	0.4904	-1.0004
0.3983	0.8062	0.3251	0.5982	0.4822	-1.0568
0.4982	0.8024	0.3642	0.5824	0.4673	-0.9934
0.5983	0.8017	0.3629	0.5691	0.4562	-0.8543
0.6985	0.8001	0.3345	0.4999	0.3999	-0.6631
0.7988	0.7990	0.2604	0.4801	0.3835	-0.4952
0.8993	0.7986	0.1402	0.3981	0.3179	-0.3001
1.0000	0.7891	0.0000	0.3462	0.2731	0.0000
T=313.15K					
0.0000	0.8670	0.0000	0.6069	0.5261	0.0000
0.0993	0.8523	0.1502	0.6049	0.5155	-0.3022
0.1989	0.8510	0.1925	0.6018	0.5129	-0.5684
0.2985	0.8442	0.2500	0.6003	0.5108	-0.8926
0.3983	0.8396	0.2890	0.6001	0.5038	-0.9534
0.4982	0.8381	0.3042	0.5982	0.5013	-0.9032
0.5983	0.8299	0.3057	0.5634	0.4675	-0.7789
0.6985	0.8200	0.2780	0.4051	0.3321	-0.5531
0.7988	0.8199	0.2139	0.3981	0.3264	-0.3852
0.8993	0.8042	0.1302	0.3553	0.2857	-0.2052
1.0000	0.7946	0.0000	0.3369	0.2677	0.0000

Table 3
Predicted kinematic viscosities and Excess molar volume For Toluene with Benzene at Different Temperatures

X_1	Y_{expt}	Y_{pred} (McAllister Model)	Y_{pred} (Jouyban- Acree Model)	Y_{pred} (K-L model)	V^E (pred) (R-K model) (cc/gmole)
T=303.15 ^o K					
0.0000	0.6242	0.6242	0.6242	0.6242	0.0000
0.0993	0.6096	0.6046	0.6021	0.6146	0.1512
0.1989	0.6057	0.6148	0.6126	0.6175	0.2413
0.2985	0.5946	0.5631	0.5413	0.6041	0.3602
0.3983	0.5185	0.5084	0.5064	0.6022	0.4320
0.4982	0.5095	0.5195	0.5394	0.6019	0.4719
0.5983	0.4947	0.4738	0.4946	0.5638	0.4701
0.6985	0.4572	0.4562	0.4661	0.5062	0.4414
0.7988	0.4197	0.4097	0.4225	0.5097	0.3217
0.8993	0.3946	0.3836	0.3926	0.4816	0.1817
1.0000	0.3559	0.3559	0.3559	0.3559	0.0000
T=308.15 ^o K					
0.0000	0.6150	0.6150	0.6150	0.6150	0.0000
0.0993	0.6049	0.6088	0.6147	0.6149	0.1223
0.1989	0.6012	0.6022	0.6134	0.6144	0.2125
0.2985	0.6001	0.6019	0.6072	0.6001	0.2704
0.3983	0.5982	0.5928	0.5984	0.5992	0.3361
0.4982	0.5824	0.5919	0.5920	0.5934	0.3652
0.5983	0.5691	0.5492	0.5592	0.5602	0.3638
0.6985	0.4999	0.4826	0.4937	0.4988	0.3354
0.7988	0.4801	0.4800	0.4820	0.4917	0.2624
0.8993	0.3981	0.3880	0.3896	0.3916	0.1422
1.0000	0.7891	0.0000	0.3462	0.2731	0.0000
T=313.15 ^o K					
0.0000	0.6069	0.6069	0.6069	0.6069	0.0000
0.0993	0.6049	0.6036	0.6053	0.6067	0.1164
0.1989	0.6018	0.6009	0.6034	0.6048	0.1993
0.2985	0.6003	0.5933	0.6023	0.6031	0.2501
0.3983	0.6001	0.5989	0.6011	0.6021	0.2843
0.4982	0.5982	0.5943	0.5992	0.5998	0.3159
0.5983	0.5634	0.5522	0.5722	0.5801	0.3257
0.6985	0.4051	0.4032	0.4077	0.4161	0.2880
0.7988	0.3981	0.3971	0.3991	0.3997	0.2243
0.8993	0.3553	0.3541	0.3561	0.3667	0.1412
1.0000	0.3369	0.3369	0.3369	0.3369	0.0000

Table 4
Parameters of McAllister constants for Toluene with Benzene System at 303.15K, 308.15K and 313.15K

Temperature T/K	v_{12}	v_{21}	SD
303.15	0.7241	0.8078	0.0147
308.15	0.7631	0.8389	0.0151
313.15	0.7209	0.7924	0.0290

Table 5
Parameters of the Krishnan and Laddha Constants and standard deviations S for the viscosity of Toluene with Benzene

Temperature T/K	A_1	A_2	A_3	SD
303.15	0.5491	-0.5473	-0.4009	0.0742
308.15	-0.5870	-3.4905	0.0847	0.0925
313.15	-3.4309	-3.6552	0.3185	0.0893

Table 6
Parameters of the Jouyban Acree model Constants and standard deviations S for the viscosity of Toluene with Benzene

Temperature T/K	A ₁	A ₂	A ₃	A ₄	SD
303.15	220.78	-200.2	69.411	204.23	0.0241
308.15	166.32	-165.2	-8.8144	162.36	0.0092
313.15	66.835	-180.24	211.710	170.870	0.0238

Table 7
Parameters of the Redlich Kister Constants and standard deviations S for Excess Volume of Toluene with Benzene

Temperature T/K	A ₁	A ₂	A ₃	A ₄	A ₅	SD
303.15	9.6517	-6.1198	-3.2991	0.6017	-2.384	0.2054
308.15	12.146	-8.3465	6.7449	4.5275	-6.771	0.3527
313.15	8.8687	-5.6724	-4.7875	2.6406	-1.6153	0.1259

Table 8
Parameters of the Redlich - Kister Constants and standard deviations S for Viscosity Deviation of Toluene with Benzene

Temperature T/K	A ₁	A ₂	A ₃	A ₄	A ₅	SD
303.15	2.1796	1.8961	-2.7342	-1.0669	3.8154	0.0422
308.15	1.7517	1.3880	-0.9378	-0.8378	0.9029	0.0069
313.15	1.9146	1.8600	-1.6801	-1.7100	2.7891	0.0488

CONCLUSION

The McAllister model, Krishnan and Laddha model and Jouyban Acree model are considered one of the best for correlating viscosity with compositions of aromatics. The models were used to correlate the kinematic viscosity –composition data for the binary systems, Toluene with benzene, 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 Toluene with benzene 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 Toluene with benzene 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 Toluene with benzene are much larger.

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CONFLICT OF INTEREST

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

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