



Statistical Approach for the Study of Viscous Behaviour of Ternary Liquid System

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ABSTRACT: The viscosity of four ternary liquid mixtures, containing two common components, chloroform and cyclohexane have been evaluated adopting statistical theory of Flory and Bloom field & Dewan theory. Molecular interaction in the system is studied in the light of excess viscosity. The agreement between theoretical and experimental values is fairly good.

Key words: Binary liquid mixtures, ternary liquid mixtures, statistical theory, excess viscosity, molecular interaction, entropy, thermal expansion coefficient, isothermal compressibility, reduced volume, equimolecular composition.

I. INTRODUCTION

Flory statistical theory [1-2] was developed and applied by various workers [3-5] for studying the viscous behaviour of binary liquid mixtures. Very few attempts [6-7] has been made to use Flory theory for study, the viscous behaviour of ternary liquid mixtures.

The present investigation, aims to evaluate theoretically, the viscosity of four ternary liquid mixtures using Flory statistical theory and Bloom field & Dewan theory. The results are compared with the experimental findings. An interaction study in the mixtures is discussed in the light of excess viscosity. The ternary mixtures under the present study are:

Benzene + Chloroform + Cyclohexane (system I)

Toluene + Chloroform + Cyclohexane (system II)

Chlorobenzene + Chloroform + Cyclohexane (system III) and

Dioxane + Chloroform + Cyclohexane (system IV)

II. THEORETICAL

Bloom field and Dewan developed a relation between viscosity and thermodynamic properties on the basis of absolute reaction rate theory and free volume theory of liquid state. Considering the possibility of two body interactions, the statistical mechanical approach of Flory [1-2] has been used to develop an expression for the dynamic viscosity of ternary liquid mixtures; the following expression for ternary liquid mixture has been obtained.

$$\ln \eta_m = \frac{-\Delta G_m^R}{RT} + f(\tilde{V}) + X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_3 \ln \eta_3 \dots (1)$$

Where $f(\tilde{V})$ is given by:

$$f(\tilde{V}) = \frac{1}{(\tilde{V}-1)} - \left[\frac{X_1}{(\tilde{V}_1-1)} + \frac{X_2}{(\tilde{V}_2-1)} + \frac{X_3}{(\tilde{V}_3-1)} \right]$$

and excess viscosity can be calculated by the expression:

$$\Delta \ln \eta = \frac{-\Delta G_m^R}{RT} + \frac{1}{(\tilde{V}-1)} - \left[\frac{X_1}{(\tilde{V}_1-1)} + \frac{X_2}{(\tilde{V}_2-1)} + \frac{X_3}{(\tilde{V}_3-1)} \right] \dots (2)$$

This residual free energy of mixing (ΔG_m^R) is generally expressed by the relation:

$$\Delta G_M^R = \Delta H_M^R - T \Delta S_M^R \dots (3)$$

Here (ΔH_M^R) is the enthalpy of mixing per mole and for ternary liquid mixture is given as:

$$\Delta H_M^R = \sum_{i=1}^3 X_i P_1^* V_1^* \left(\frac{1}{(\tilde{V}_i)} - \frac{1}{(\tilde{V})} \right) + \frac{X_1 V_1^* \theta_2 X_{(1,2)}}{\tilde{V}_1} + \frac{X_2 V_2^* \theta_3 X_{(2,3)}}{\tilde{V}_2} + \frac{X_3 V_3^* \theta_1 X_{(3,1)}}{\tilde{V}_3} \dots (4)$$

Viscous behaviour cannot be explained solely on the basis of free volume effects but residual entropy of mixing has been considered [26] to act as major additional factor of activation flow.

The residual entropy of mixing (ΔS_m^R) can be evaluated using the relation:

$$\Delta S_M^R = \sum_{i=1}^3 \frac{3X_i P_i^* V^*}{T_1^*} \ln \frac{(\tilde{V}_1^{1/3} - 1)}{(\tilde{V}^{1/3} - 1)} \quad ..(5)$$

Combining equations (1) to (4) the expression for viscosity of ternary liquid mixture takes the form:

$$\begin{aligned} \ln \eta_{(m)} = & [X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_3 \ln \eta_3] - X_1 P_1^* V_1^* \left[\left(\frac{1}{\tilde{V}_1} - \frac{1}{\tilde{V}} \right) + 3\tilde{T}_1 \ln \frac{\tilde{V}_1^{1/3} - 1}{\tilde{V}^{1/3} - 1} \right] \\ & - X_2 P_2^* V_2^* \left[\left(\frac{1}{\tilde{V}_2} - \frac{1}{\tilde{V}} \right) + 3\tilde{T}_2 \ln \frac{(\tilde{V}_2^{1/3} - 1)}{(\tilde{V}^{1/3} - 1)} \right] - X_3 P_3^* V_3^* \left[\left(\frac{1}{\tilde{V}_3} - \frac{1}{\tilde{V}} \right) \right] \\ & + 3\tilde{T}_3 \ln \frac{(\tilde{V}_3^{1-3} - 1)}{(\tilde{V}^{1-3} - 1)} + \frac{X_1 V_1^* \theta_2 X_{(1,2)}}{\tilde{V}_1} + \frac{X_2 V_2^* \theta_2 X_{(2,3)}}{\tilde{V}_2} + \frac{X_3 V_3^* \theta_1 X_{(3,1)}}{\tilde{V}_3} \\ & \left[\frac{1}{(\tilde{V} - 1)} - \left(\frac{X_1}{(\tilde{V}_1 - 1)} + \frac{X_2}{(\tilde{V}_2 - 1)} + \frac{X_3}{(\tilde{V}_3 - 1)} \right) \right] \quad ..(6) \end{aligned}$$

The characteristics pressure, temperature and reduced volume for the liquids are evaluated using the following equations:

$$\begin{aligned} P^* &= \frac{\alpha T (\tilde{V})^2}{\beta_T} \\ T^* &= \frac{T (\tilde{V})^{4/3}}{[(\tilde{V})^{1/3} - 1]} \\ \tilde{V} &= \left(\frac{\alpha T}{3(1 + \alpha T)} + 1 \right)^3 \quad ..(7) \end{aligned}$$

Where β_T , T and β_T represents the thermal expansion coefficient, temperature and isothermal compressibility respectively.

The reduced volume for mixture is given by:

$$v = \frac{V}{X_1 V_1^* + X_2 V_2^* + X_3 V_3^*} \quad ..(8)$$

The segment and site fractions of the ternary mixtures are calculated using following relations [19]

$$\Psi_3 = \frac{X_3}{X_3 + X_1 \left(\frac{V_1^*}{V_2^*} \right) + X_2 \left(\frac{V_2^*}{V_3^*} \right)} \quad ..(9)$$

$$\Psi_2 = \frac{X_2}{X_2 + X_3 \left(\frac{V_3^*}{V_2^*} \right) + X_1 \left(\frac{V_1^*}{V_2^*} \right)} \quad ..(10)$$

$$\text{and } \Psi_1 = 1 - (\Psi_2 + \Psi_3)$$

$$\theta_3 = \frac{\Psi_3}{\Psi_3 + \Psi_1 \left(\frac{V_3^*}{V_1^*} \right)^{1/3} + \Psi_1 \left(\frac{V_3^*}{V_2^*} \right)^{1/3}} \quad ..(11)$$

$$\theta_2 = \frac{\Psi_2}{\Psi_2 + \Psi_3 \left(\frac{V_2^*}{V_3^*} \right)^{1/3} + \Psi_1 \left(\frac{V_2^*}{V_1^*} \right)^{1/3}} \quad ..(12)$$

$$\text{and } \theta_1 = 1 - (\theta_2 + \theta_3)$$

The interaction parameters are obtained using the relations:

$$X_{1,2} = P_1^* \left[1 - \left(\frac{P_2^*}{P_1^*} \right) \left(\frac{S_1}{S_2} \right)^{1/2} \right]^2 \quad ..(13)$$

$$X_{2,3} = P_2^* \left[1 - \left(\frac{P_3^*}{P_2^*} \right) \left(\frac{S_2}{S_3} \right)^{1/2} \right]^2 \quad ..(14)$$

$$X_{3,1} = P_3^* \left[1 - \left(\frac{P_1^*}{P_3^*} \right) \left(\frac{S_3}{S_1} \right)^{1/2} \right]^2 \quad ..(15)$$

Bingham proposed for binary liquid mixture which gives the ideal viscosity of the mixture:

$$\eta = X_1\eta_1 + X_2\eta_2 \quad \dots(16)$$

For n-component mixture eq. (16) can be rewritten as:

$$\eta = \sum_{i=1}^n X_i\eta_i \quad \dots(17)$$

Where X_i and η_i are respectively the mole fraction and viscosity of pure components.

Following Kendall and Munroe, the viscosity for multi component systems can be evaluated by the relation:

$$\ln\eta = \sum_{i=1}^n X_i \ln\eta_i \quad \dots(18)$$

$$\ln\eta = X_1^2 \ln\eta_1 + X_2^2 \ln\eta_2 + X_3^2 \ln\eta_3 + 2(X_1X_2 \ln X_1^2 \ln\eta_{1,2} + X_2X_3 \ln X_1^2 \ln\eta_{2,3} + X_3X_1 \ln\eta_{3,1}) + 3X_1X_2X_3 \ln\eta_{1,2,3} \quad \dots(20)$$

For ternary liquid mixtures, the equation suggested by, Hind *et al.* takes the form:

$$\eta = X_1^2\eta_1 + X_2^2\eta_2 + X_3^2\eta_3 + 2[X_1X_2\eta_{(1,2)} + X_2X_3\eta_{(2,3)} + X_3X_1\eta_{(3,1)}] + 3X_1X_2X_3\eta_{1,2,3} \quad \dots(21)$$

In the present investigation, a different approach has been adopted to obtain the constant $\eta_{(1,2)}$, $\eta_{(2,3)}$, $\eta_{(3,1)}$ and $\eta_{(1,2,3)}$. These equations are obtained from the respective pure components of the mixtures taking their equimolecular composition and following additively rule. Thus:

$$\eta_{i,j} = 0.5\eta_i + 0.5\eta_j \quad (22)$$

Similarly $\eta_{(1,2,3)}$ is obtained from pure components as.

$$\eta_{(1,2,3)} = \frac{1}{3}\eta_1 + \frac{1}{3}\eta_2 + \frac{1}{3}\eta_3 \quad \dots(23)$$

III. RESULTS AND DISCUSSIONS

Application of the Flory as well as Bloom field and Dewan theory for the evaluation of viscosity and the study of the ternary liquid mixtures: Benzene + Chloroform + Cyclohexane (I) Toluene + Chloroform + Cyclohexane (II) Chlorobenzene + Chloroform + Cyclohexane (III) Dioxane + Chloroform + Cyclohexane (IV)

The characteristic pressure (P^*) reduced temperature (\bar{T}) and reduced volume (\bar{V}) of pure components are calculated using eqs. (7) for the above ternary systems.

The values of reduced volume (\bar{V}) and the site fraction of the mixture at different mole fraction and the values of interaction energy parameter are obtained vide eqs. (8), (11), (12), (13), (14) and (15) respectively.

The values of (\bar{V}), (\bar{T}) and (V^*) and (P^*) of the pure components and reduced volume (\bar{V}) of the mixture and the values of energy parameter have been employed to obtain the values of excess free energy of mixing at composition vide eqs. (3).

The viscosity of ternary mixtures under consideration is computed through Bloom field & Dewan and Flory theory vide equation (6). The computed viscosity along with their percentage deviation from experimental values are presented in Table 1. The excess viscosity has been

Bingham and Kendall relations were proposed considering the ideal mixing of solutions which is not always true.

Taking into consideration the interaction between the molecules, Frankel with the help of Eyring's model, developed the logarithmic relation for non ideal binary liquid mixtures, given as:

$$\ln\eta = X_1^2 \ln\eta_1 + X_2^2 \ln\eta_2 + 2X_1X_2 \ln \dots(19)$$

Where $\eta_{1,2}$ is a constant.

This equation 19 when extended to ternary liquid mixture takes the following term:

obtained using equation (2) and are also incorporated in table 1. The necessary data, required for the calculation have been taken from the literature.

A perusal of table 1 shows that there is good agreement between theoretical and experimental values of viscosity showing an average percentage deviation of 3.94 for the system (I), 4.04 for the system (II) 2.16 for the system (III) and 4.29 for the system (IV) respectively. The trend of computed values of viscosities also agrees with the experimental values for all the systems under consideration.

The theoretical excess viscosities (η^E) has been evaluated using eq. 2 and are also incorporated in table 1 along with experimental excess viscosities are found to be positive over the whole composition range for all the four systems under consideration. These values are in good agreement with the values of experimental excess viscosities. The sign and magnitude of (η^E) may be visualized in terms of strength of interactions operating between the components of any system. According to Fort and Moore, the positive values of excess viscosity (η^E) are indication of weak interaction while negative values shows strong interaction between unlike molecules.

In the present work, the four ternary systems containing two common components, chloroform and cyclohexane have been considered. The variation of (η^E) with composition of the mixtures suggests the presence of interaction between the components. The positive values of (η^E) in the systems shows the presence of weak interactions.

Due to the non polar nature and inertness of cyclohexane strong interaction with cyclohexane is not expected. Therefore, only dispersive forces are dominant between benzene toluene chlorobenzene and cyclohexane.

Due to presence of C-H dipole in chloroform, it can interact considerably with the electron donating molecules such as benzene, toluene, chlorobenzene and dioxane forming a donor acceptor complex in the case of chloroform with benzene or toluene and chlorobenzene and weak hydrogen bonding in chloroform with dioxane.

It is clear from the table that percentage deviation for these systems are : 3.94%, 4.04%, 2.175% and 4.30% respectively, for the system I, II, III & IV due to Pandey et al and the deviation due to present approach are -9.17%, -6.08, 4.3 and -5.95% respectively. Thus it can be concluded that the present approach satisfactory gives the values of viscosity by Frankel relation.

Table 1: Statistical Approach for the Study of Viscous Behaviour of ternary liquid systems.

x_1	x_2	G_m^R RT	$I_\eta \eta_v$	$\sum X_1 l_n \eta$	$I_n \eta(c.p.)$	$\eta(c.p.)$		% Deviation	η^E	
				Ideal visco (C.P.)	Theoretical	Theoretical	Experimental		Theoretical	Experimental

SYSTEM I Benzene (1) + Chloroform (2) + Cyclohexane (3)

0.1015	0.5966	0.01187	-0.03696	-0.33850	-0.38665	0.67930	0.62900	-8.00%	0.95300	0.88200
0.2019	0.4991	0.01665	-0.02240	-0.37015	-0.40919	0.66420	0.60160	-10.40%	0.96200	0.87100
0.2998	0.4009	0.02025	-0.09896	-0.40187	-0.52108	0.59390	0.58210	-2.00%	0.88700	0.87000
0.4001	0.2997	0.20907	-0.08687	-0.43455	-0.54232	0.58140	0.57230	-1.60%	0.89800	0.88400
0.5045	0.1972	0.01845	-0.07603	-0.46776	-0.56224	0.56990	0.65380	-1.08%	0.90900	0.90000
0.6031	0.0993	0.01433	-8.06597	-0.49944	-0.57974	0.56000	0.56320	+0.56%	0.92300	0.92800
Average % deviation Δ								3.94%		

SYSTEM II Toluene (1) + Chloroform (2) + Cyclohexane (3).

0.0911	0.5404	0.00460	-0.06103	-0.36093	-0.42655	0.65270	0.61670	-5.84	0.93640	0.88470
0.2198	0.4863	0.00556	-0.05892	-0.39171	-0.45620	0.63368	0.60210	-5.25	0.93750	0.89080
0.3004	0.3960	0.00600	-0.04291	-0.42680	-0.47571	0.62144	0.58930	-5.45	0.95220	0.90300
0.4013	0.2961	0.00696	-0.08687	-0.46703	-0.56086	0.57072	0.57080	-4.21	0.91040	0.91056
0.5016	0.1985	0.00650	-0.02178	-0.50655	-0.53433	0.58577	0.56210	-4.21	0.97210	0.93280
0.5976	0.0989	0.00631	+0.01005	-0.54611	-0.54236	0.58137	0.56180	-3.48	1.00370	0.96990
Average % deviation Δ								4.04%		

SYSTEM III Chlorobenzene (1) + Chloroform (2) + Cyclohexane (3)

0.1001	0.5965	0.00965	-0.08313	-0.31654	-0.31654	0.66410	0.5472	-2.61%	0.91139	0.88820
0.1988	0.4978	0.01433	-0.10428	-0.32682	-0.32682	0.64055	0.6455	+0.77%	0.88810	0.89502
0.3150	0.3975	0.01705	-0.08851	-0.33448	-0.33448	0.64402	0.6453	+0.20%	0.89980	0.90162
0.4005	0.3001	0.01693	-0.00942	-0.34672	-0.34672	0.68860	0.6504	-5.87%	0.97400	0.91993
0.5023	0.1995	0.01633	-0.03679	-0.35698	-0.35698	0.66860	0.6625	-0.16%	0.94820	0.94672
0.5997	0.0996	0.01422	-0.04650	-0.36783	-0.36783	0.65145	0.6740	+3.34%	0.94108	0.97365
Average % deviation Δ								+2.17%		

SYSTEM IV Dioxane (1) + Chloroform (2) + Cyclohexane (3).

0.1023	0.5979	0.02090	-0.02890	-0.27580	-0.32550	0.72215	0.69130	-4.46%	0.95150	0.91080
0.2024	0.4976	0.02594	-0.10010	-0.24710	-0.37320	0.68860	0.71810	+4.11%	0.88160	0.91940
0.2981	0.4008	0.03030	-0.05610	-0.22000	-0.30640	0.73610	0.75270	+2.21%	0.91720	0.93790
0.3991	0.3097	0.03714	-0.11160	-0.18830	-0.33700	0.71390	0.77560	+7.96%	0.86180	0.93630
0.5001	0.1992	0.02990	-0.01380	-0.16200	-0.20560	0.81420	0.82110	+0.84%	0.95730	0.96550
0.6046	0.0986	0.02484	-0.00310	-0.13080	-0.15880	0.85310	0.87530	+2.54%	0.97240	0.99770
Average % deviation Δ								4.30%		

Table 2: Theoretical Viscosities for ternary liquid system (in C.P.).

X ₁	X ₂	Bengham I	Kendall II	Frenkel III	Hind IV	Experimental	% Δ (I)	% Δ (II)	% Δ (III)	% Δ (IV)
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SYSTEM I Benzene(1) + Chloroform (2) + Cyclohexane (3)

0.1015	0.3019	0.7200	0.7128	0.7005	0.7564	0.6290	-14.46%	-13.30%	-11.37%	-20.26%
0.2019	0.2990	0.6984	0.6906	0.669	0.7582	0.6016	-16.07%	-14.79%	-11.22%	-26.03%
0.2998	0.2993	0.6766	0.6690	0.6435	0.7481	0.5821	-16.23%	-14.92%	-10.55%	-28.52%
0.4001	0.3002	0.6542	0.6475	0.6225	0.7258	0.5723	-14.30%	-13.13%	-8.77%	-26.82%
0.5045	0.2983	0.6315	0.6264	0.6062	0.6905	0.5638	-12.00%	-11.90%	-7.51%	-22.47%
0.6031	0.2976	0.6098	0.6068	0.5950	0.5831	0.5632	-8.27%	-7.74%	-5.64%	-3.53%
Average % deviation Δ							-13.56%	-12.63%	-9.17%	-21.27%

SYSTEM II Toluene (1) + Chloroform (2) + Cyclohexane (3)

0.0911	0.3685	0.7053	0.6970	0.6849	0.4315	0.6167	-14.37%	-13.02%	-11.04%	+30.03%
0.2198	0.2939	0.6858	0.6759	0.6536	0.4994	0.6021	-13.90%	-12.26%	-8.55%	+17.06%
0.3004	0.3036	0.6625	0.6526	0.6273	0.5262	0.5893	-12.42%	-10.74%	-6.57%	+10.71%
0.4013	0.3026	0.6360	0.6269	0.6024	0.5170	0.5708	-11.42%	-9.83%	-5.53%	+10.53%
0.5016	0.2999	0.6099	0.6026	0.5831	0.4662	0.5621	-8.50%	-7.21%	-3.74%	+17.06%
0.5976	0.3035	0.5838	0.5792	0.5679	0.3750	0.5618	-3.92%	-3.10%	-1.08%	+33.25%
Average % deviation Δ							-10.76%	-9.36%	-6.08%	+19.77%

SYSTEM IV Chlorobenzene (1) + Chloroform (2) + Cyclohexane (3)

0.1001	0.3034	0.7343	0.7287	0.7180	0.7728	0.6472	-13.46%	-12.59%	-10.94%	-19.41%
0.1988	0.3034	0.7264	0.7212	0.7019	0.7907	0.6455	-12.53%	-11.73%	-8.73%	-22.49%
0.3150	0.2875	0.7202	0.7157	0.6920	0.7970	0.6453	-11.61%	-10.91%	-7.24%	-23.51%
0.4005	0.2994	0.7112	0.7070	0.6834	0.7879	0.6504	-9.35%	-8.70%	-5.07%	-21.14%
0.5023	0.2982	0.7034	0.6998	0.6804	0.7670	0.6625	-6.17%	-5.63%	-2.70%	-15.77%
0.5997	0.3007	0.6952	0.6922	0.6810	0.7334	0.6740	-3.15%	-2.70%	-1.04%	-8.81%
Average % deviation Δ							-9.38%	-8.71%	-5.95%	-18.52%

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