Transport Properties of Oxygenated Ternary Liquid Mixture at Temperature 298.15 K

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Abstract: Density and viscosity were measured for some oxygenated ternary system at 298.15K and atmospheric pressure and further compared with the theoretical results obtained from two models (Bertrand-Acree- Bruchfield and Flory). The properties were fitted to the Redlich-Kister polynomial equation to estimate the binary coefficients and standard errors. The deviation in viscosity was also computed to study the nature and extent of the molecular interactions in the ternary mixtures. Testing of the models for the ternary system showed that a fair agreement is achieved between experimental and theoretical results when they are compared. Conclusively, both theoretical models were consistent with the experimental results.

Keywords: Bertrand- Acree-Bruchfield , Flory model , Redlich-Kister, Ternary and Viscosity

I. Introduction

Some oxygenated compounds are usually added to gasoline to improve the octane number and reduce pollution. The present work is concerned with the oxygenated compounds of the type either cyclic ether or aliphatic alcohol and the alkane liquid that generally appears in gasoline. From the viewpoint of association, cyclic ethers can be regarded as an intermediate case between alkanes (inert compounds) and alkanols (highly self-associated compounds).

Viscosity is an important thermo physical property used in industrial technology to study the mass transfer in a rotating packed bed with viscous newtonian and non-newtonian fluids¹, breakage parameter of quartz², liquid-solid fluidization³, diffusion and the mesoscopic hydrodynamics of super cooled liquids⁴, in the use of nano liter viscometer for analyzing blood plazma and other liquid samples⁵, shear viscosity relaxation of liquid n-alkanes⁶, in the flow of viscous shear thinning fluids behind cooling coil banks in large reactors⁷, on shear and longitudinal behaviour of non-ionic $c_{12}e_8$ aqueous solution⁸.

For these reasons, we measured densities, viscosities of ternary and binary mixtures of tetrahydrofuran, 2-propanol, and 2,2,4- trimethylpentane at temperature 298.15 K and atmospheric pressure. Present study involves the experimental and theoretical study (using the predictive model of Bertrand- Acree-Bruchfield and Flory theory), of viscosity for ternary liquid mixtures: Tetrahydrofuran + 2-Propanol + 2,2,4-Trimethylpentane and their contributory binaries: (1)Tetrahydrofuran + 2-Propanol), (2) Tetrahydrofuran + 2,2,4-Trimethylpentane and (3) 2-Propanol + 2,2,4-Trimethylpentane binary systems. Deviation in viscosity, $\Delta \eta$ were evaluated and fitted to the Redlich-Kister polynomial⁹ to derive the binary coefficients and standard errors.

II. Models

2.1 Bertrand-Acree-Bruchfield Model:

Bertrand, Acree and coworkers¹⁰⁻¹³ have developed a predictive equation, based on a model ternary system for the studies of the thermo-chemical properties of solute in simple binary solvents. This equation obeys the general mixing equation,

$$\Delta Z_{123}^{-ex} = (n_1 \Gamma_1 + n_2 \Gamma_2 + n_3 \Gamma_3)^{-1} (n_1 \Gamma_1 n_2 \Gamma_2 A_{12} + (n_1 \Gamma_1 n_3 \Gamma_3 A_{13} + n_2 \Gamma_2 n_3 \Gamma_3 A_{23})$$
(1)

Where Z_{ij} represents any extensive thermodynamic property described in the terms of interaction parameters A_{ij} and weighting factors Γ_i . This equation leads to a very good predictive form, for combining the properties of binary systems.For for the multicomponent system,

$$\Delta Z_{1,2...N}^{-ex} = \sum_{i=1}^{N} \sum_{j>1}^{N} (X_i + X_j)(f_i + f_j)(\Delta Z_{ij}^{-ex})^*$$
(2)

However a method for the calculation of the ratio of raw weighting factors has been given by Bertrand et al⁸³. Here ratio is calculated for each binary combination of the components of the multicomponent system, which are then combined to normalize the weighting factors to an average value of 100 by,

$$\Gamma_{i} = (100N)[(\Gamma_{1}^{raw}/\Gamma_{i}^{raw}) + (\Gamma_{2}^{raw}/\Gamma_{i}^{raw}) + \dots + (\Gamma_{N}^{raw}/\Gamma_{i}^{raw})$$
(3)

If we reasonably modify this approach in the light of the above findings then $\phi_1, \phi_2 \dots \phi_n$ can be used in the place of weighted mole fractions, and the equations takes up the following form:

$$\Delta Z_{1,2...,N}^{E} = \sum_{i=1}^{N} \sum_{j>1}^{N} (X_{i} + X_{j})(\phi_{i} + \phi_{j})(\Delta Z_{ij}^{E})$$

(4)

Where X_i and X_j are the mole fractions of multicomponent system, and ϕ_i are ϕ_j are their respective volume fractions and (ΔZ^E_{ij}) can be more appropriately taken as the experimental excess property of the constituent binary combinations at the same molar ratio of the components as in the multicomponent system.

Results so obtained were compared with the experimental findings, and with the results of PFP which utilize the properties of single components to predict the thermodynamic properties of multicomponent liquid mixtures.

2.2 Flory theory:

Mecedo and Litovitz¹⁴ made the hypothesis that the two effects (absolute rate theory and free volume theory) are combined so that the probabilities for viscous flow is taken as the product of the probabilities for acquiring sufficient activation energy and of the occurrence of an empty site. A bridge can be formed to the thermodynamic functions of mixing by assuming a simple relationship between solution activation energy ΔG^{\pm} , the pure liquid activation energy ΔG_1^{\pm} , and the residual Gibbs free energy of mixing ΔG_M^R .

$$\Delta G^{\neq} = x_1 \Delta G_1^{\neq} + x_2 \Delta G_2^{\neq} - \Delta G_M^R$$

(5)

where x_1 and x_2 are mole fraction of the component.

The values of ΔG_{M}^{R} was evaluated from Flory theory and may be expressed as;

$$\Delta G_{M}^{R} = x_{1} P_{1}^{*} V_{1}^{*} \left[\left(\frac{1}{\tilde{V}_{1}} - \frac{1}{\tilde{V}} \right) + 3\tilde{T}_{1} \ln \frac{(\tilde{V}_{1}^{1/3} - 1)}{(\tilde{V}^{1/3} - 1)} \right] + x_{2} P_{2}^{*} V_{2}^{*} \left[\left(\frac{1}{\tilde{V}_{2}} - \frac{1}{\tilde{V}} \right) + 3\tilde{T}_{2} \ln \frac{(\tilde{V}_{2}^{1/3} - 1)}{(\tilde{V}^{1/3} - 1)} \right] + \frac{X_{1} P_{1}^{*} V_{1}^{*} \theta_{2} X_{12}}{\tilde{V}_{1}} \right]$$
(6)

By combining the Eyring¹⁵ relation for the residual Gibbs free energy of mixing, ΔG^R_M and $\Delta G^{\#}$, we have obtained the following expression for the viscosity η of a binary liquid system:

$$RT \ln \eta = \sum_{i=1}^{2} x_{i} \ln \eta_{i} - \left[\sum_{i=1}^{2} x_{i} P_{i}^{*} v_{i}^{*} \left\{ (1/v_{i}^{-} - 1/v) + \frac{1}{3T} \ln(\frac{v_{i}^{-1/3}}{(v_{i}^{-} - 1)}) - \frac{1}{(v_{i}^{-} - 1)} \right\} \\ = \sum_{i=1}^{2} \sum_{j=1}^{2} (x_{i} v_{i}^{*} \theta_{j} x_{ij} / v_{i}) / RT \left(\frac{1}{v_{i}^{-} - 1} \right) - \left(\sum_{i=1}^{2} \frac{x_{i}}{v_{i}^{-} - 1} \right)$$

(7)

Here, P^* , v^* , v, T, θ and X_{ij} are the characteristic pressure, characteristic volume, reduced volume, reduced temperature, site fraction and interaction parameter respectively. All the notations in the above equation have their usual significance as detailed out by Flory¹⁶⁻¹⁷.

III. Tables and Figures

Table1.Thermal expansion coefficient (α), isothermal compressibility(β_T),molar volume (V), density (ρ) and viscosity (η) of pure components for comparisons at 298.15.

Components	Density / $\rho_{(g.cm^3)}$		Viscosity / η(mPa·s)		
	Exp	Lit*	Exp	Lit*	
tetrahydrofuran	0.8823	0.8829	0.470	0.530	
2-propanol	0.7811	0.7812	2.043	2.0436	
2,2,4-trimethyl pentane	0.6879	0.6878	0.481	0.4802	

*Ref.[18]

Table 2. Binary Coefficients of the Redlich–Kister Equation and Standard Deviations ($\Delta \sigma$) of η for the Binary Systems at 298.15 K

	A0	A1	A2	A3	$\Box \sigma$			
Tetrahydrofuran + 2-Propanol								
$\Delta \eta / mPa \cdot s$ -2.3927 1.5854 -0.9562 0.4833					3.4			
Tetrahydrofuran +2,2,4-Trimethylpentane								
Δη/mPa·s -0.0104 0.0097 -0.0001 -0.0215 0.03								
2-Propanol +2,2,4-Trimethylpentane								
$\Delta \eta / mPa \cdot s$	-2.4618	-1.3794	-0.7282	-0.7792	2.4			

Table 3. Experimental densities (ρ_{exp}) viscosities (η_{exp}) viscosity deviation $(\Delta \eta)$, theoretical viscosities (η_{Flory}) and percent deviation $(\%\eta_{Flory})$ for binary systems at 298.15 K

x_1	ρ_{exp}	η_{exp}	$\Delta \eta$	η_{Flory}	% _{<i>η</i>_{Flory} mPa}					
	$g \cdot cm^{-3}$	mPa · s	mPa · s	mPa · s	•\$					
Tetra hydrofuran + 2-Propanol										
0.0500	0.78631	1.706	-0.258	1.796	5.27					
0.0500	0.78631	1.706	-0.258	1.796	5.27					
0.1000	0.79135	1.445	-0.440	1.512	4.63					
0.1500	0.79635	1.248	-0.559	1.314	5.29					
0.2001	0.80135	1.113	-0.615	1.176	5.66					
0.2500	0.80634	0.988	-0.662	1.008	2.02					
0.3000	0.81133	0.896	-0.675	0.904	8.92					
0.3500	0.81633	0.812	-0.680	0.856	5.42					
0.4000	0.82131	0.752	-0.662	0.793	5.45					
0.4500	0.82630	0.703	-0.632	0.762	8.39					
0.5000	0.83129	0.659	-0.598	0.701	6.37					
0.5500	0.83632	0.623	-0.555	0.688	10.43					
0.6000	0.84135	0.594	-0.506	0.623	4.88					
0.6500	0.84639	0.567	-0.453	0.607	7.05					
0.7000	0.85145	0.545	-0.397	0.586	7.52					
0.7500	0.85652	0.526	-0.337	0.552	4.94					
0.8000	0.86166	0.511	-0.274	0.532	4.11					
0.8500	0.86681	0.497	-0.209	0.516	3.82					
0.9000	0.87202	0.488	-0.139	0.503	3.07					
0.9500	0.87721	0.480	-0.069	0.493	2.71					
	Tetra	ahydrofuran + 2,	2,4-Trimethylpen	tane	1					
0.0500	0.69268	0.481	-0.701	0.501	4.15					
0.1001	0.69770	0.479	-0.912	0.499	4.17					
0.1500	0.70302	0.479	-1.212	0.499	4.17					
0.2000	0.70866	0.477	-1.841	0.496	3.39					
0.2500	0.71465	0.476	-2.133	0.495	3.99					
0.3000	0.72104	0.475	-2.724	0.494	4.00					

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0.3500	0.72787	0.475	-2.516	0.493	3,79					
0.4000	0.73518	0.474	-2.607	0.491	3.58					
0.4500	0.74300	0.473	-2.616	0.491	3.80					
0.5000	0.75136	0.473	-2.551	0.489	3.38					
0.5500	0.76031	0.473	-2.524	0.482	1.90					
0.6000	0.76992	0.472	-2.416	0.480	1.69					
0.6500	0.78026	0.472	-2.304	0.479	1.48					
0.7000	0.79141	0.472	-1.337	0.478	1.27					
0.7500	0.80347	0.471	-1.306	0.479	1.69					
0.8000	0.81655	0.471	-1.216	0.479	1.69					
0.8500	0.83080	0.471	-1.231	0.478	1.48					
0.9000	0.84638	0.471	-0.125	0.477	1.27					
0.9500	0.86349	0.470	-1.048	0.476	1.27					
	Propanol + 2.2.4-Trimethylpentane									
0.0500	0.68936	0.482	-0.077	0.496	2.90					
0.1000	0.69113	0.486	-0.150	0.499	2.67					
0.1501	0.69320	0.493	-0.221	0.503	2.03					
0.2000	0.69548	0.503	-0.290	0.516	2.58					
0.2500	0.69796	0.515	-0.356	0.523	1.55					
0.3000	0.70066	0.530	-0.419	0.531	0.02					
0.3500	0.70364	0.550	-0.477	0.567	3.09					
0.4000	0.70690	0.576	-0.529	0.582	1.04					
0.4500	0.71045	0.608	-0.576	0.615	1.15					
0.5000	0.71431	0.646	-0.615	0.670	3.71					
0.5500	0.71851	0.694	-0.645	0.704	1.44					
0.6000	0.72309	0.751	-0.666	0.767	2.13					
0.6500	0.72812	0.819	-0.677	0.833	1.71					
0.7000	0.73362	0.901	-0.673	0.981	8.87					
0.7500	0.73968	1.003	-0.649	1.023	0.19					
0.8000	0.74632	1.122	-0.608	1.196	6.59					
0.8500	0.75369	1.277	-0.532	1.304	2.11					
0.9000	0.76183	1.407	-0.417	1.485	5.54					
0.9500	0.77098	1.707	-0.258	1.796	5.21					

Volume – 03, *Issue* – 06, *June* 2018, *PP* – 01-07

Table 4. Experimental densities (ρ_{exp}), viscosities (η_{exp}), viscosity deviation($\Delta \eta$), theoretical viscosities (η_{Flory} , η_{BAB}) and their percentage deviations ($\Delta \% \eta$) for the Tetrahydrofuran + 2-Propanol +2,2,4-Trimethylpentane at 298.15 K

<i>x</i> ₁	x_2	ρ g·cm ⁻³	η_{Exp} mPa·s	$\Delta \eta$ mPa·s	η _{Flory} mPa·s	η _{BAB} mPa·s	$\Delta \% \eta_{Flory}$	$\Delta \% \eta_{BOB}$
0.0500	0.9000	0.7758	1.467	-0.419	1.487	1.443	1.36	1.63
0.0501	0.7999	0.7580	1.122	-0.608	1.142	1.100	1.78	1.96
0.0500	0.7000	0.7436	1.905	-0.669	1.925	1.880	1.5	1.31
0.0500	0.6000	0.7318	0.759	-0.658	0.781	0.737	2.89	2.89
0.0500	0.5001	0.7220	0.655	-0.606	0.675	0.636	3.05	2.90
0.0500	0.4000	0.7138	0.586	-0.519	0.606	0.568	3.41	3.07
0.0500	0.3000	0.7069	0.541	-0.407	0.562	0.521	3.88	3.69
0.0500	0.2000	0.7011	0.509	-0.283	0.529	0.491	3.92	3.53
0.0500	0.1000	0.6965	0.491	-0.144	0.511	0.501	4.07	2.03
0.0500	0.0500	0.6944	0.484	-0.073	0.504	0.489	4.13	1.03
0.1000	0.8500	0.7806	1.256	-0.552	1.276	1.231	1.59	1.99
0.1000	0.7500	0.7623	0.998	-0.653	1.018	0.976	2.00	2.20
0.1000	0.6500	0.7477	0.821	-0.674	0.836	0.800	1.82	2.55
0.1000	0.5500	0.7358	0.701	-0.638	0.721	0.680	2.85	2.99
0.1000	0.4500	0.7258	0.622	-0.560	0.642	0.605	3.21	2.73

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0.1000	0.3499	0.7174	0.561	-0.465	0.583	0.545	3.92	2.85
0.1000	0.2500	0.7103	0.523	-0.347	0.536	0.510	2.48	2.48
0.1000	0.1501	0.7045	0.499	-0.214	0.520	0.488	4.20	2.40
0.1000	0.0500	0.6997	0.483	-0.074	0.503	0.471	4.14	2.48
0.2000	0.7500	0.7901	0.991	-0.659	1.011	0.969	2.01	2.21
0.2000	0.6500	0.7712	0.814	-0.680	0.826	0.794	1.47	2.45
0.2000	0.5500	0.7561	0.698	-0.640	0.718	0.681	2.86	2.43
0.2000	0.4500	0.7437	0.617	-0.564	0.637	0.601	3.24	2.59
0.2000	0.3500	0.7334	0.562	-0.463	0.583	0.545	3.73	3.02
0.2000	0.2500	0.7246	0.523	-0.346	0.543	0.508	3.82	2.86
0.2000	0.1501	0.7174	0.498	-0.214	0.507	0.481	1.80	3.41
0.2000	0.0500	0.7112	0.482	-0.074	0.491	0.469	1.86	2.69
0.3000	0.6500	0.7995	0.814	-0.679	0.834	0.794	2.45	2.45
0.3000	0.5500	0.7801	0.699	-0.638	0.719	0.679	2.86	2.86
0.3000	0.4500	0.7645	0.621	-0.559	0.644	0.601	3.70	3.22
0.3000	0.3500	0.7516	0.565	-0.459	0.585	0.554	3.53	1.94
0.3001	0.2500	0.7409	0.526	-0.342	0.546	0.510	3.80	3.04
0.3000	0.1500	0.7320	0.508	-0.203	0.528	0.491	3.93	3.34
0.3000	0.0500	0.7243	0.488	-0.067	0.498	0.471	2.04	3.48
0.4000	0.5500	0.8091	0.704	-0.632	0.724	0.681	2.84	3.26
0.4000	0.4500	0.7891	0.621	-0.558	0.642	0.601	3.38	3.22
0.4000	0.3500	0.7729	0.565	-0.458	0.585	0.548	3.53	3.00
0.3999	0.2501	0.7596	0.533	-0.334	0.554	0.520	3.93	2.43
0.4000	0.1500	0.7484	0.499	-0.211	0.519	0.481	4.00	3.60
0.4000	0.0500	0.7393	0.482	-0.072	0.492	0.471	2.07	2.28
0.5000	0.4500	0.8187	0.623	-0.556	0.644	0.601	3.37	3.53
0.5000	0.3500	0.7981	0.566	-0.456	0.586	0.551	3.53	2.65
0.5000	0.2500	0.7814	0.531	-0.335	0.541	0.515	1.88	3.01
0.5000	0.1501	0.7677	0.501	-0.208	0.521	0.484	3.99	3.39
0.5000	0.0500	0.7564	0.482	-0.071	0.502	0.469	4.14	2.69
0.6000	0.3500	0.8284	0.570	-0.451	0.591	0.550	3.68	3.58
0.6000	0.2500	0.8077	0.527	-0.338	0.547	0.509	3.79	3.41
0.6000	0.1500	0.7901	0.498	-0.211	0.512	0.480	2.81	3.61
0.6000	0.0500	0.7761	0.479	-0.073	0.491	0.461	2.50	3.75
0.7000	0.2500	0.8385	0.528	-0.336	0.538	0.511	1.89	3.21
0.7000	0.1500	0.8165	0.499	-0.209	0.513	0.481	2.80	3.60
0.6999	0.0500	0.7990	0.481	-0.070	0.501	0.466	4.15	3.11
0.8000	0.1500	0.8484	0.503	-0.204	0.513	0.488	1.98	2.98
0.8000	0.0500	0.8260	0.482	-0.068	0.497	0.470	3.11	2.48
0.8999	0.0501	0.8585	0.481	-0.068	0.496	0.469	3.11	2.49

Volume – 03, *Issue* – 06, *June* 2018, *PP* – 01-07



Figure 1 Viscosity deviation for the ternary mixtures formed by addition of 2,2,4 Trimethylpentane to the binary mixtures of Tetrahydrofuran + 2-Propanol at 298.15 K. Experimental results, ●; Flory model,O;BAB model, ▲.



Figure 2. Viscosity deviation for the binary mixtures vs mole fraction *x*. Experimental results: •, Tetra hydrofuran + 2-Propanol; O, Tetrahydrofuran + 2,2,4-Trimethylpentane and ; \blacktriangle , Propanol + 2,2,4-Trimethylpentane.

IV. Results and Discussion

Experimental values of density and viscosities for the pure liquids at 298.15K were compared with those found in the literature¹⁸ and were found to be in fair agreement, as shown in TABLE 1.The ternary mixtures data at 298.15 K for tetrahydrofuran, 2-propanol, and 2,2,4- trimethylpentane are presented in TABLE 4. TABLE 3 list the values of theoretical viscosity, viscosity deviations ($\Delta\eta$), for the contributory binaries from the ternary mixtures at 298.15 K.

The viscosity deviation $(\Delta \eta)$ were represented mathematically by the Redlich-Kister equation for correlating the experimental data,

$$y = x_1(1-x_1)\sum_{i=0}^{p} A_i (2x_1-1)^i$$

where (y) refers to $(\Delta \eta)$, x_i is the mole fraction and A_i is the coefficients. The values of coefficients A_i were determined by a multiple regression analysis based on the least squares method and are summarized along with the standard deviations between the experimental and fitted values of the respective function in TABLE 2. The standard deviation is defined by.

$$\sigma = \left[\sum_{i=1}^{m} (y_{\exp_i} - y_{cal_i})^2 / (m-p)\right]^{1/2}$$

(9)

(8)

where (m) is the number of experimental points and (p) is the number of adjustable parameters. For the case the highest value of $(\Delta \sigma)$ corresponds to 2-propanol+ 2,2,4-trimethylpentane while the lowest value of $(\Delta \sigma)$ corresponds to tetrahydrofuran+2,2,4-trimethylpentane. The values of the viscosity deviations, $\Delta \eta$, for the mixtures are calculated from the viscosity values of the pure liquids, η_i , and their mixtures, η , from the equation as:

$$\Delta \eta = \eta - \sum_{i=1}^{N} x_i \eta_i$$

(10)

between unlike molecules and negative contributions. In the present investigation, tetrahydrofuran is associated through the dipole–dipole interaction and 2-propanol is associated through the hydrogen bonding of its hydroxyl group. It is observed that in the binary liquid mixtures, $\Delta \eta$ values are negative over the whole mole fractions. The values of $\Delta \eta$ are negligibly small for tetrahydrofuran + 2,2,4-trimethylpentane mixtures. The values of $\Delta \eta$ at 298.15 K vary from –0.680 mPa · s to –0.001 mPa · s. The viscosity deviation $\Delta \eta$ (*x*) 0.5 increases in the order: tetrahydrofuran + 2-propanol \approx 2-propanol + 2,2,4-trimethylpentane < tetrahydrofuran + 2,2,4-trimethylpentane < 0. The values of $\Delta \eta$ increase with a rise in composition except for those of the tetrahydrofuran + 2,2,4-trimethylpentane system. A careful perusal of TABLE 4 indicate the

variation of the values of density, viscosity with the mole fraction of the component. The regular trend observed in TABLE 4 reveals the dominance of like interactions. With the addition of third component (2,2,4-Trimethylpentane) in the mixture, density and viscosity of ternary mixture decreases where as deviation in viscosity increases up to some extent then increases with negative values indicating strong interactions are prevailing in the mixture as evident in Fig. 1 and 2.

V. Conclusion

Prediction of the thermodynamic and transport properties of liquid mixture depends largely on the extent of interactions involved in the liquid mixture. A good correlation between the theoretical and experimental values clearly point out towards the consideration of all the possible interactions present. The magnitude of such properties gives the measure of various interactions which may be associated with a particular system. Structural chemistry is related to the interactions involved in the liquid systems. Nature and type of bonding associated with the structure can readily be studied through the study of interactions.

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