Volumetric and Viscometric behavior studies for ternary system of cycloheptane with three isomeric dimethyl benzene at different temperatures.

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Abstract:

This study concerned with measurement the densities ρ and viscosities η. Over the range of temperatures (293.15, 303.15, 313.15, and 323.15 k) for ternary mixtures cycloheptane + m-xylene + o-xylene and cycloheptane + m-xylene + p-xylene. From experimental data of densities and viscosities, the excess molar volumes, V^E, and excess viscosities, η^E , were calculated for ternary mixtures. The Flory theory has been extended for the theoretical prediction of excess molar volume of ternary mixtures studied here depending on the pure component liquid parameters. In this study Heric-Coursey equation was used to calculate the excess viscosities for ternary mixtures studied here.

Key words: Ternary system, Cyclohptane , dimethyl benzene isomer, Volumetric and Viscometric of Ternary system.

Introduction:

Dimethyl benzene or xylene is one of the top 30 chemicals produced in the United states in terms of volume. It is used as a solvent in the printing, rubber, and leather industries. Along with other solvents, xylene is also used as a cleaning agent, a thinner for paint, and in varnishes[1]. It is found in small amounts in airplane fuel and gasoline. xylene is used as a material in chemical, plastics, and synthetic fiber industries and as an ingredient in the coating of fabrics and papers. Isomers of xylene are used in the manufacture of certain polymers, such as plastics[2].

Many industrial processes use mixtures of solvents and knowledge of chemical and physical properties of pure liquids and their mixtures is critical to the efficient utilization-preparation development and economic design of appropriate equipment for these processes are only possible if the chemical and physical properties of the solvent or substances to be processed are known. For this reason, many attemps have been made to correlate these data using semi-empirical or theoretical approaches considerable programs have also been made in the development of statistical theories. The calculation of physical properties using statistical theories is still not yet possible for complicated molecules such as xylene and long chain molecules, because of the range of different conformations, which can occur and the effect of these various structures on the intermoleculer interactions[3]. Recently, few equations for estimating the excess thermodynamic and other physicochemical properties of multicomponent systems from the observed properties of their various contributory of the components has been developed and applied to various multicomponent liquid systems[4,5] computer-stimulated calculations and neutron-scattering experiments[6] have been insight into the relations between molecular structure and macroscopic behavior considerable progress has also been made in the development statistical theories over recent years[7].

Experimental:

(a) Materials: All the chemicals used were supplied by BDH and Fluka. Company.

The purities of all substances were better than 99 mass% as found by GLC analysis.

The purity of the chemicals was checked by comparing the densities and viscosities of the components with those reported in the literature[12].

(b) Measurements:

(1) Densities measurement:- Densities were measured at 293.15, 303.15, 313.15, and 323.15 k with an Antopaar digital densimeter (Model DMA 60/601) Densities were measured with a precision of 2×10^{-5} g.cm^{-3}. The maximum uncertainty in the excess molar volumes is expected to be less than 3×10^{-3} cm^3.mol^{-1}.

(2) Viscosities Measurements:- In the present work, the viscosity (η) of the solution was measured using (cannon-ubbelohde semi micro) viscometer. The temperature of the

solution was brought to the desired value by immersing the test viscometer in a controlled temperature water bath with a precision of ± 0.01k

Results:

Experimental results of the densities ρ_m of the ternary mixtures cycloheptane + m-xylene + o-xylene and cycloheptane + m-xylene + p-xylene at (293.15, 303.15, 313.15, and 323.15k) are listed in Table (1).

The excess molar volumes for ternary mixtures studied here were calculated from the measured, densities using the following equation:-

$$V_{123}^{E}(cm^3.mol^{-1}) = \left[\frac{X_1M_1 + X_2M_2 + X_3M_3}{\rho_m}\right] - \left[X_1\frac{M_1}{\rho_1} + X_2\frac{M_2}{\rho_2} + X_3\frac{M_3}{\rho_3}\right] \quad \text{.....(1)}$$

Where $X_{(1,2,3)}$, $M_{(1,2,3)}$ and $\rho_{(1,2,3)}$ are respectively the mole fraction, molar mass and density of the pure component liquid (1,2,3), $\rho_{(m)}$ is the density mixture. The obtained results of V_{123}^{E} are listed in table (3) and a plotted as a function of the mole fraction X_1, X_2 , and X_3 for the three components at four temperatures in figures (1 and 2).

The statistical mechanical concept of Flory theory[13] has been extended for the theoretical prediction of excess molar volume of the ternary mixture from the properties of pure component. The excess molar volumes (V^E) calculated directly from characteristic and reduced volumes and the segment fraction using thermal expansion coefficient (α) of the pure three component, and using the equation.

$$V^{E} = (X_1V_1^* + X_2V_2^* + X_3V_3^*)[\tilde{V} - (\phi_1\tilde{V}_1 + \phi_2\tilde{V}_2 + \phi_3\tilde{V}_3)] \quad \text{.....(2)}$$

Where ϕ_1, ϕ_2 and ϕ_3 are the segment fractions of components 1, 2, and 3 defined by the relations:-

$$\phi_1 = [1 - \phi_2 - \phi_3] \quad \text{.....(3)}$$

$$\phi_2 = \frac{X_2}{X_2 + X_3\left(\frac{V_3^*}{V_2^*}\right) + X_1\left(\frac{V_1^*}{V_2^*}\right)} \quad \text{.....(4)}$$

$$\phi_3 = \frac{X_3}{X_3 + X_2\left(\frac{V_2^*}{V_3^*}\right) + X_1\left(\frac{V_1^*}{V_3^*}\right)} \quad \text{.....(5)}$$

$\tilde{V}$ in equation (6) is the reduced volume of ternary mixture which is obtained by the following equation:-

$$\tilde{V}=V/X_1V_1^* + X_2V_2^* + X_3V_3^* \quad .....(6)$$

Where V is the molar volume of the mixture, given by:-

$$V = (X_1M_1 + X_2M_2 + X_3M_3)/\rho_m \quad .....(7)$$

Where ρ_m is the density of the mixture.

By using the equation of state parameters of pure liquids, table (2) and applied equation (2-7), we calculated the excess molar volumes for the ternary mixtures studied here. Table (3) present the theoretical prediction of V^E values at 293.15k with experimental values for comparison for ternary mixtures studied here. The maximum percent average deviation is less than 0.95% which means that Flory theory for predicting the excess molar volumes of ternary mixtures studied here is quite reasonable, evident from this excellent agreement in both sign and magnitude.

The absolute viscosity (η_m) was calculated for ternary mixtures in this study by measuring the flow time (τ) and using the densities values shown in table (1) at the range of degrees of temperature (293.15, 303.15, 313.15 and 323.15 k) and by using equation (8).

$$\eta = c.\tau.\rho_m \quad .....(8)$$

Where (c) the viscosity constant

Table (4) presents the experimental values of absolute viscosities for ternary mixtures studied here at the range of degrees of temperature.

Excess viscosities, $\eta^{E}_{1,2,3}$ for ternary mixtures at the range of degrees of temperature were calculated from measurements of the viscosity of the mixture and the pure liquid by the following equation:

$$\eta^{E}_{1,2,3} = \eta_{mix.} - X_1\eta_1 - X_2\eta_2 - X_3\eta_3 \quad \text{.....(9)}$$

Where X_1, X_2, X_3 , η_1. η_2 , and η_3 are the mole fractions and the viscosities of components 1,2, and 3 respectively, η_{mix} is the viscosity of the mixture.

The obtained result of $\eta^{E}_{1,2,3}$ are listed in table (5) and plotted as a function of the mole fractions X_1, X_2, and X_3 for the three components at four temperatures in figures (3 and 4).

Several empirical equations have been proposed to calculate excess viscosity of multi component systems based on the available experimental results of viscosities of mixture and pure components, the equation of Heric and coursey [14] was used:

$$\eta^{E} = \ln \eta_{mix} - \sum_{xi} xi \ln \eta_i \quad \text{.....(10)}$$

Where η_{mix} is the viscosity of the mixture, X_i and η_i are mole fraction and viscosity for pure component respectively. Table (5) shows the experimental result of $\eta^{E}_{1,2,3}$ and the predicted values of excess viscosity from equation [10] for comparison for ternary mixtures studied here at (293.15, 303.15, 313.15 and 323.15k).

Discussion:

The experimental excess molar volume $V^{E}_{1,2,3}$ for the ternary mixtures studied here at 393.15, 303.15, 313.15 and 323.14k are positive deviation from ideality over the whole mole fraction, table (3) and figures (1 and 2). Such volumetric behavior may be explained by the globular molecules of cycloheptane disturb the orientational order in isomers of xylene. It means that cycloheptane molecules are interstitial accommodate between isomers of xylene molecules and result less packed structure which is responsible about the positive $V^{E}_{1,2,3}$ for ternary mixtures studied here.

The Flory theory [13] has been extended for the theoretical prediction of excess molar volume of ternary mixtures studied here depending on the pure component liquid parameters, the obtained excess molar volumes $V^{E}_{1,2,3}$ by extended Flory theory for the ternary mixtures studied here at 293.15k are presented in table (3) with the experimental data for comparison. The theory predicted the volumetric behavior and magnitudes of $V^{E}_{1,2,3}$ well.

The researcher conclude that Flory theory could be extended to multi component liquid mixtures based on the pure component liquid parameters[15].

Experimental data of mixture viscosity η_m and excess viscosity $\eta^E_{1,2,3}$ are listed in tables (4 and 5) and a plotted as a function of the mole fractions X_1, X_2, and X_3 for the three components at four temperature in figures (3 and 4) for the ternary mixture studied here. The viscometric behavior for the ternary mixture studied here show a similar volumetric behavior. Heric and Coursey equation was used to calculated excess viscosity of ternary mixtures studied here from experimental results of viscosities of mixtures and pure components. Table (5) shows good agreement between the experimental values of the excess viscosities for the ternary mixtures studied in this work with the predicted values from Heric and Coursey equation.

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دراسة السلوك الحجمي واللزوجي للنظام الثلاثي المكون من السايكلوهبتان مع الايزومرات الثلاثة للداي مثيل بنزين عند درجات حرارية مختلفة

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الخلاصة:

تتضمن هذه الدراسة قياس الكثافة واللزوجة على مدى اربع درجات حرارية للمخاليط الثلاثية المكون والتي تتضمن :
cycloheptane + m-xylene + o-xylene and cycloheptane + m-xylene + p-xylene.
ومن النتائج العملية لقياس الكثافة واللزوجة تم حساب الحجوم المولارية الفائضة V^E ، واللزوجة الفائضة η^E للمخاليط الثلاثية قيد الدراسة.
تم تطبيق نظرية فلوري لحساب الحجوم المولارية الفائضة للمخاليط الثلاثية المكون. هذه الدراسة استخدمت معادلة (Heric-Coursey) لحساب اللزوجة للمخاليط الثلاثية المكون.

Table (1) Experimental values of the densities (ρ) for ternary mixtures at four temperatures.

X1 cycloheptane + x2 m-xylene + x3 o-xylene					
X1	X2	ρ ($g.cm^{-3}$) 293.15k	ρ ($g.cm^{-3}$) 303.15k	ρ ($g.cm^{-3}$) 313.15k	ρ ($g.cm^{-3}$) 323.15k
0.2848	0.3715	0.7898	0.7687	0.7501	0.7412
0.2860	0.3534	0.7576	0.734	0.7239	0.7125
0.2909	0.3395	0.7155	0.7025	0.7004	0.6988
0.2923	0.3214	0.6913	0.6832	0.67	0.6667
0.3097	0.3080	0.6609	0.6415	0.6346	0.6223
0.3271	0.2928	0.6412	0.6227	0.6052	0.5974
0.3460	0.2718	0.6334	0.5944	0.5838	0.5781

0.3679	0.2440	0.6147	0.5863	0.5742	0.5509
0.3846	0.2225	0.585	0.5574	0.5459	0.5345
0.3960	0.2155	0.5539	0.5385	0.511	0.5037
0.4286	0.2088	0.5422	0.5293	0.5003	0.4889
0.4302	0.1942	0.5319	0.5011	0.4871	0.479

X1 cycloheptane + x2 m-xylene + x3 p-xylene .I					
X1	X2	ρ ($g.cm^{-3}$) 293.15k	ρ ($g.cm^{-3}$) 303.15k	ρ ($g.cm^{-3}$) 313.15k	ρ ($g.cm^{-3}$) 323.15k
0.2739	0.3585	0.77321	0.74984	0.72089	0.71977
0.2741	0.3209	0.74564	0.72531	0.70785	0.6951
0.2869	0.3035	0.71083	0.69158	0.68665	0.67083
0.2890	0.2700	0.68547	0.67484	0.66807	0.65312
0.2970	0.2412	0.65331	0.63097	0.62515	0.61774
0.3091	0.2278	0.63402	0.60553	0.5819	0.57002
0.3265	0.2100	0.59984	0.58568	0.56571	0.55519
0.3498	0.1624	0.57039	0.56001	0.53432	0.52132
0.3769	0.1363	0.56445	0.54131	0.51009	0.49748
0.4000	0.1200	0.54387	0.5222	0.49781	0.46312
0.4244	0.0885	0.52334	0.49921	0.47321	0.45078
0.4789	0.0563	0.50792	0.4751	0.45112	0.43662

Table (2) Parameters for the pure liquids according to the Flory Theory at 298.15k.[8-11]

Liquid	$V/cm^3.mol^{-1}$	$V^*/cm^3.mol^{-1}$	$\tilde{V}$	T^*/K	$\tilde{T}$	$P^*/J.cm^{-3}$ *	$\alpha \times 10^{-3}/k^{-1}$ *	S/A^{o-1*}
cycloheptane	119.4511	95.2462	1.2542	4942.07	0.1658	٥٤٨	1.109	0.89
o- xylene	123.4552	76.311	1.6178	3312.10	0.1111	٣٧٠	0.588	0.79
m-xylene	123.7427	99.207	1.2473	5276.02	0.1770	٣٨٨	0.998	0.82
p-xylene	124.1725	111.839	1.1103	9822.03	0.3294	٣٦٧	٠.392	0.78

Table (3) Experimental and theoretical prediction values of the (V^E_{123}) for ternary mixtures at four temperatures.

X1 cycloheptane + x2 m-xylene + x3 o-xylene						
X1	**X2**	$V^E_{123}\,exp.$ **($cm^3.mol^{-1}$) 293.15k**	$V^E_{123}pred.$ **($cm^3.mol^{-1}$) 293.15k**	V^E_{123} **($cm^3.mol^{-1}$) 303.15k**	V^E_{123} **($cm^3.mol^{-1}$) 313.15k**	V^E_{123} **($cm^3.mol^{-1}$) 323.15k**
0.2848	0.3715	0.1826	0.1832	0.2149	0.2112	0.1800
0.2860	0.3534	0.2943	0.3117	0.3428	0.3124	0.2931
0.2909	0.3395	0.4551	0.4367	0.4691	0.4087	0.3497
0.2923	0.3214	0.5566	0.5719	0.5527	0.5441	0.4928
0.3097	0.3080	0.6920	0.6628	0.7471	0.7141	0.7122
0.3271	0.2928	0.7856	0.7638	0.8415	0.8700	0.8480
0.3460	0.2718	0.8223	0.8554	0.9961	0.9923	0.6904
0.3679	0.2440	0.9178	0.9640	1.0399	1.0481	1.1328

0.3846	0.2225	1.0853	1.1147	1.2189	1.2316	1.2445
0.3960	0.2155	1.2808	1.1825	1.3461	1.4870	1.4777
0.4286	0.2088	1.3541	1.3359	1.4046	1.5644	1.5936
0.4302	0.1942	1.4273	1.4713	1.6229	1.6762	1.6803

X1 cycloheptane + x2 m-xylene + x3 p-xylene .II						
X1	X2	V^{E}_{123} exp. (cm^3.mol^{-1}) 293.15k	V^{E}_{123} *pred.* (cm^3.mol^{-1}) 293.15k	V^{E}_{123} (cm^3.mol^{-1}) 303.15k	V^{E}_{123} (cm^3.mol^{-1}) 313.15k	V^{E}_{123} (cm^3.mol^{-1}) 323.15k
0.2739	0.3585	0.2374	0.2380	0.2822	0.3222	0.3272
0.2741	0.3209	0.3374	0.3269	0.3769	0.3782	0.3432
0.2869	0.3035	0.4723	0.4864	0.5149	0.4677	0.4510
0.2890	0.2700	0.5806	0.5908	0.5898	0.5540	0.5378
0.2970	0.2412	0.7289	0.7321	0.8030	0.7679	0.7212
0.3091	0.2278	0.8237	0.8327	0.9390	1.0129	1.0013
0.3265	0.2100	1.0071	0.9557	1.0517	1.1120	1.0965
0.3498	0.1624	1.1814	1.1703	1.2096	1.3247	1.3381
0.3769	0.1363	1.2145	1.2828	1.3312	1.5030	1.5240
0.4000	0.1200	1.3481	1.3419	1.4653	1.5975	1.8271
0.4244	0.0885	1.4916	1.4564	1.6412	1.8076	1.9450
0.4789	0.0563	1.5986	1.6078	1.8355	2.0056	2.0803

Table(4) Experimental values of the absolute viscosity (η) for ternary mixtures at four temperatures.

X1 cycloheptane + x2 m-xylene + x3 o-xylene					
X1	X2	η (293.15k)	η(303.15k)	η(313.15k)	η (323.15k)
0.2848	0.3715	1.9531	1.8743	1.6911	1.5423
0.2860	0.3534	1.9295	1.8535	1.6674	1.5209
0.2909	0.3395	1.8982	1.8323	1.6587	1.5018
0.2923	0.3214	1.8843	1.8099	1.6367	1.4929
0.3097	0.3080	1.8522	1.7891	1.621	1.4741
0.3271	0.2928	1.8313	1.7603	1.6048	1.4557
0.3460	0.2718	1.8051	1.7594	1.5889	1.4369
0.3679	0.2440	1.7836	1.7345	1.5753	1.4178
0.3846	0.2225	1.7678	1.7254	1.5559	1.4083
0.3960	0.2155	1.7463	1.7042	1.5483	1.3992
0.4286	0.2088	1.7259	1.6873	1.5348	1.37
0.4302	0.1942	1.7047	1.6775	1.5055	1.3534

X1 cycloheptane + x2 m-xylene + x3 p-xylene					
X1	X2	η (293.15k)	η (303.15k)	η(313.15k)	η (323.15k)
0.2739	0.3585	1.8952	1.8697	1.6132	1.5224
0.2741	0.3209	1.8713	1.8442	1.5959	1.5119
0.2869	0.3035	1.8601	1.8251	1.5715	1.5007
0.2890	0.2700	1.8419	1.7936	1.5543	1.4892
0.2970	0.2412	1.8278	1.7789	1.5112	1.4712
0.3091	0.2278	1.8159	1.7542	1.5034	1.4534
0.3265	0.2100	1.7986	1.7364	1.4987	1.4397
0.3498	0.1624	1.7791	1.7119	1.4772	1.4229
0.3769	0.1363	1.7556	1.6828	1.4583	1.4101
0.4000	0.1200	1.7384	1.6709	1.4211	1.4074
0.4244	0.0885	1.7002	1.6667	1.4102	1.3998
0.4789	0.0563	1.6887	1.6583	1.4032	1.3902

Table(5) : Experimental and the predicted excess viscosities (η^E) for ternary mixtures at 29٣.15k, 30٣.15k, 313.15k,and 323.15k.

(Cycloheptane + x_2 m- xylene + x_3 o- xylene)									
x_1	x_2	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$
		293.15k		303.15k		313.15k		323.15k	
0.2848	0.3715	0.8488	0.8485	0.8115	0.8112	0.6934	0.6931	0.6062	0.6058
0.2860	0.3534	0.8233	0.8217	0.7893	0.7890	0.6686	0.6682	0.5836	0.5832
0.2909	0.3395	0.7858	0.7843	0.7627	0.7622	0.6551	0.6544	0.5602	0.5601
0.2923	0.3214	0.7698	0.7699	0.7386	0.7376	0.6318	0.6315	0.5500	0.5511
0.3097	0.3080	0.7162	0.7154	0.6976	0.6954	0.5984	0.5974	0.5156	0.5153
0.3271	0.2928	0.6738	0.6733	0.6487	0.6477	0.5646	0.5632	0.4816	0.4810
0.3460	0.2718	0.6242	0.6231	0.6259	0.6251	0.5295	0.5299	0.4458	0.4453
0.3679	0.2440	0.5756	0.5755	0.5758	0.5745	0.4939	0.4930	0.4072	0.4069
0.3846	0.2225	0.5391	0.5372	0.5474	0.5470	0.4576	0.4569	0.3827	0.3825
0.3960	0.2155	0.5036	0.5021	0.5130	0.5124	0.4384	0.4377	0.3634	0.3624
0.4286	0.2088	0.4434	0.4447	0.4582	0.4589	0.3917	0.3911	0.3051	0.3047
0.4302	0.1942	0.4199	0.4186	0.4466	0.4461	0.3609	0.3613	0.2870	0.2873

(Cycloheptane + x_2 m- xylene + x_3 p- xylene)									
x_1	x_2	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$	$\eta^E_{exp.}$	$\eta^E_{Pred.}$
		293.15k		303.15k		313.15k		323.15k	
0.2739	0.3585	0.8054	0.8052	0.3631	0.3625	0.6333	0.6328	0.5995	0.5991
0.2741	0.3209	0.7802	0.7811	0.3099	0.3092	0.6154	0.6151	0.5883	0.5879
0.2869	0.3035	0.7530	0.7527	0.2632	0.2627	0.5778	0.5775	0.5655	0.5652
0.2890	0.2700	0.7313	0.7315	0.2048	0.2041	0.5581	0.5579	0.5517	0.5511
0.2970	0.2412	0.7068	0.7062	0.1598	0.1590	0.5065	0.5054	0.5261	0.5260
0.3091	0.2278	0.6799	0.6785	0.1113	0.1117	0.4862	0.4853	0.4974	0.4968
0.3265	0.2100	0.6409	0.6401	0.0602	0.0609	0.4635	0.4631	0.4679	0.4680
0.3498	0.1624	0.5918	0.5913	-0.0261	-0.0263	0.4178	0.4175	0.4297	0.4291
0.3769	0.1363	0.5348	0.5339	-0.1057	-0.1055	0.3710	0.3716	0.3924	0.3927
0.4000	0.1200	0.4891	0.4892	-0.1563	-0.1558	0.3100	0.3108	0.3689	0.3691
0.4244	0.0885	0.4205	0.4208	-0.2119	-0.2109	0.2739	0.2733	0.3391	0.3388
0.4789	0.0563	0.3420	0.3417	-0.3071	-0.3062	0.2110	0.2107	0.2805	0.2803