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Thermo Physical Behavior of Binary and Ternary System's For Cyclohexanol With Some Ketones at Three Temperatures.

السلوك الثرموفيزيائي للنظام الثنائي والثلاثي للسايكلو هكسانول مع بعض الكيتونات عند ثلاث درجات حرارية

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Abstract:

The density and viscosity of binary mixtures cyclohexanol + 2-hexanone, cyclohexanol + 2-heptanone and ternary mixture cyclohexanol + 2-hexanone + 2-heptanone have been measured over the range of temperatures (293.15K, 303.15K and 313.15K).

The excess molar volume and excess viscosity of binary systems were also calculated, and the results were fitted to a well-known Redlich-kister-type polynomial equation. As for the ternary system studied, the excess molar volume shows a positive from ideal at the three different temperatures studied. The excess viscosity shows positive deviation from the ideality at (293.15K and 303.15K) at decrease mole fraction X_1, and at the temperature (313.15K) shows a positive deviation and becomes too negative deviation at increase of the mole fraction X_1.

Keyword: Binary and ternary systems, cyclohexanol , 2-hexanone, 2-heptanone, excess molar volume ,density ,viscosity

الخلاصة:

تم دراسة الخواص الفيزيائية من كثافة ولزوجة للنظام الثنائي للمزيج المكون من cyclohexanol + 2-heptanone, والنظام الثلاثي للمزيج المكون من cyclohexanol + 2-hexanone + 2-heptanon cyclohexanol + 2-hexanone عند درجات الحرارة (293.15K, 303.15K and 313.15K) وتم قياس الحجم المولاري الفائض واللزوجة الفائضة للأنظمة الثنائية المدروسة. ومن حساب النتائج وجدت مطابقة إلى معادلة Redlich-Kister .

أما بالنسبة للنظام الثلاثي المدروس فأن الحجم المولاري الفائض يظهر انحرافاً موجباً عن المثالية عند درجات الحرارة الثلاثة عند (293.15K and 303.15K)المدروسة. أما فائض اللزوجة تظهر انحرافاً موجباً عند الدرجتين الحراريتين X_1. تظهر انحرافاً موجباً يتحول إلى الانحراف السالب بزيادة الكسر المولي 313.15K وعند درجة حرارةX_1انخفاض الكسر المولي

Introduction:

The practical studies of binary and ternary mixtures reveal the importance of molecular interactions (hydrogen bonding, charge-transfer complexes, dipole-dipole, dipole-induced dipole, interstitial accommodate chain alignment …. etc) on the physical properties of these mixtures[1]. Knowledge of the mixing properties are useful in design and simulation processes, in the synthesis of pharmaceuticals, lacquers, resins, polymers, oxygenated fuels, and paint[2,3], some of these properties are density and viscosity. Several solvent parameters are frequently employed to investigate and interpret medium effects in chemical reactions and other solution processes, using for instance linear salvation energy relationships analysis[4-6]. Moreover, selected properties of

solvents are required for the control of industrial chemical processes, such as polymerization and solvolysis. The density and viscosity are between the most relevant intensive properties of solvents.

Recently, extensive studies of thermo mechanical properties and solvatochromic parameters of binary alcohol mixtures were undertaken [7-11] in order to improve our understanding on salvation phenomena.

However, investigations of the physical properties of 2-hexanone and 2-heptanone in a binary mixture with alcohol, especially cyclohexanol, over a wide range of temperatures and compositions are still scarce and not available in the literature.

Therefore, this study aimed to obtain reliable density and viscosity data for binary and ternary mixtures of these ketones at (293.15K, 303.15K, and 313.15K).

Experimental (Materials):

All chemicals were supplied by Fluka and Aldrich chemical company Inc. and used without any further purification. The purity of these materials is (99%) for cyclohexanol, (99.9%) for 2-hexanone, and (99.95%) for 2- heptanone.

The binary and ternary mixture samples were prepared by mass in airtight-stoppered glass bottles using a single pan mettler balance (Sartorius) with an uncertainty of $\pm 10^{-4}$g. The uncertainty of the mole fraction for each binary and ternary mixture is less than 0.0001.

Density Measurements:

Measurements of the densities of the pure components, the binary and ternary mixtures were carried out using a Anton paar digital densimeter (model DMA 60/602) with an uncertainty of about 10^{-5}g.cm^{-3}. Prior to measurement, the instrument was calibrated with double-distilled water. The temperature of the measuring cell was maintained at (293.15K, 303.15K, and 313.15K) using a (HAKKE Dl-E) with an uncertainty of 0.01K.

Viscosity Measurements:

The viscosity of the solutions was measured by using (Cannon-Ubbelohde semi Micro) viscometer. This viscometer had an approximate constant (0.02 centistokes/second), a capillary length is about (8 cm) and diameter about (0.36 mm).

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. A temperature regulation called (Temp-Unite) type (Kottermann) was used. All measurements described above were performed at least three times, and the results were averaged to give the final values.

Results and Discussion:

Table (1) summarizes the experimental results of the density and viscosity of the pure liquid and compares these results to those from literatures. The experimental values were in a good agreement with literature [12]. The density and viscosity of binary and ternary mixtures of (cyclohexanol + 2-hexanon, cyclohexanol + 2-heptanone, and cyclohexanol + 2-hexanone + 2-heptanone) obtained in this study at three different temperatures are given in tables (2 and 3).

The excess molar volume, V^{E}, and excess viscosity, η^{E}, were calculated from density and viscosity measurements according to the following equations [13-16].

$$V_{1,2}^{E} = \frac{X_1M_1+X_2M_2}{\rho_m} - (X_1V_1+X_2V_2) \qquad .....(1)$$

$$\eta_{1,2}^{E} = \eta_m - X_1\eta_1 - X_2\eta_2 \qquad .....(2)$$

for binary mixtures, and

$$V_{1,2,3}^{E} = \frac{X_1M_1+X_2M_2+X_3M_3}{\rho_m} - (X_1V_1+X_2V_2+X_3V_3) \qquad .....(3)$$

$$\eta_{1,2,3}^{E} = \eta_m - X_1\eta_1 - X_2\eta_2 - X_3\eta_3 \qquad .....(4)$$

for ternary mixture.

where, ρ_m is the density of binary and ternary mixture in eq. 1 and 3 respectively X_1, V_1, M_1, X_2, V_2, M_2 and X_3, V_3, M_3 are the mole fraction, molar volume, and molecular weight of pure components 1,2, and 3, respectively in equations 1 and 3. The terms η_m, η_1, η_2, and η_3 in eq.2 and 4 are the viscosity of the binary and ternary mixture, viscosity of pure components 1,2 and 3, respectively.

The calculated excess molar volumes for the given binary and ternary systems are also summarized in tables (2 and 3). From these results, one can see that the excess molar volumes for (X_1 cyclohexanol + X_2 2-hexanone) in this system is negative from ideality at low mole fractions of 2-hexanone $0 < X_2 < 0.5$ and becomes positive at high mole fraction, while $V_{1,2}^E$ for (X_1 cyclohexanol + X_2 2- heptanone) is positive over the whole mole fractions range at three different temperatures.

$V_{1,2}^E$ Become more and more positive with increasing the chain length of ketone and with decreasing the temperature. This suggests that the globular molecule cyclohexanol disturbing the hydrogen bonding interaction in ketones and result less packed structure. It seem that in addition to the above discussed molecular interactions, the size and the molar volume of ketones could increase the deviation from ideality.

The results of the excess molar volume for binary mixtures, $V_{1,2}^E$, as a function of the mole fraction of component, 2, X_2 at (293.15, 303.15, and 313.15)K are depicted in figures 1 and 2, In this figures, the calculated excess molar volumes, $V_{1,2}^E$ for binary systems are represented by symbols, where as the solid lines represent the well-known Redlich-Kister polynomial equation for excess molar volume, V^E, which has the form [17].

$$V_{1,2}^E = X_1 X_2 \sum_{i=0}^{n} A_i (X_1 - X_2)^i \quad .....(5)$$

Where A_i represents the parameters of the Redlich-Kister polynomial equation and those obtained from the least-squares method. The parameters of the Redlich-Kister polynomial equation for excess molar volume V^E are given in table 4.

The excess molar volume $V_{1,2,3}^E$ for ternary mixture are plotted as a function of the mole fraction X_1, X_2 and X_3 for the three components at three different temperatures in figure 3. In this figure, the excess molar $V_{1,2,3}^E$, shows a positive deviation over the whole composition range at 293.15K, 303.15K and 313.15K.

The experimental values of viscosity for binary and ternary mixtures studied in this work listed in tables (2 and 3) were used to calculate excess viscosity using eq.2 and 4.

Excess viscosities $\eta_{1,2}^E$ are negative over the entire composition range for two binary mixtures studied here at three different temperatures. The negative deviation from ideality decreases when increasing the chain length of ketones and increasing the temperature.

The excess viscosity $\eta_{1,2}^E$ for binary mixtures are plotted as a function of the mole fraction of component 2 (X_2 at 293.15, 303.15, and 313.15)K in figures 4 and 5.

In these figures, the calculated excess viscosity $\eta_{1,2}^E$ for binary systems studied here are represented by symbols, and the solid lines represent the well-known Redlich-Kister polynomial equation for the excess viscosity, which has the following form:

$$\eta_{1,2}^E = X_1 X_2 \sum_{i=0}^{n} A_i (X_1 - X_2)^i \quad .....(6)$$

The parameters A_i of eq.(6) obtained by the least-Squares method are given in table 4. From this table it is clear that the Redlich-Kister polynomial equation can represents the excess molar volume and excess viscosity very well, which is indicated by low standard deviations.

The excess viscosity $\eta^{E}_{1,2,3}$ for ternary mixture studied here are plotted as a function of the mole fraction, X_1, X_2 and X_3 for the three components at three different temperatures in figure 6. In this figure, the calculated excess viscosity Show a positive deviation over the whole composition range at 293.15K and 303.15K, but at 313.15K the excess viscosities show a positive deviation at low mole fraction X_1 of cyclohexanol and a negative deviation at high mole fraction.

Table (1): Comparison of the experimental density and viscosity of pure compounds with literature values at 293.15K, , 303.15K, and 313.15K.

Compound	T/K	$\rho(g\,cm^{-3})$		$\eta(mpa.s)$	
		exp.	**lit.**	**exp.**	**lit.**
Cyclohexanol	293.15 303.15 313.15	0.9418 0.9394 0.9254	---------- 0.9415 ---------	0.6091 0.3532 0.1925	-------- 0.4106 ---------
2-hexanone	293.15 303.15 313.15	0.8075 0.8024 0.7877	--------- 0.8022 ---------	0.0054 0.0048 0.0042	--------- --------- ---------
2-heptanone	293.15 303.15 313.15	0.8134 0.8066 0.7928	--------- 0.8067 ---------	0.0073 0.0064 0.0057	--------- 0.0068 ---------

Table (2): Experimental values of density and viscosity and calculated values of excess molar volume and excess viscosity for binary mixtures of studied here at (293.15, 303.15, and 313.15)K.

X_1cyclohexanol + X_2 2-hexanone												
X_2	ρ(g.cm^{-3}) 293.15K	V^E cm^{3}mol^{-1} 293.15K	η 293.15K	η^Emp.s 293.15K	ρ(g.cm^{-3}) 303.15K	V^E cm^{3}mol^{-1} 303.15K	η 303.15K	η^Emp.s 303.15K	ρ(g.cm^{-3}) 313.15K	V^E cm^{3}mol^{-1} 313.15K	η 313.15K	η^Emp.s 313.15K
0.0000	0.94182	0.00000	0.60916	0.00000	0.93940	0.00000	0.35321	0.00000	0.92548	0.00000	0.19250	0.00000
0.0862	0.92852	-0.00104	0.24550	-0.31163	0.92658	-0.09374	0.15181	-0.17137	0.91206	-0.03880	0.08190	-0.09437
0.1796	0.91501	-0.06000	0.13350	-0.36724	0.91200	-0.06561	0.08171	-0.20892	0.89752	-0.02720	0.05851	-0.10017
0.2778	0.90106	-0.10186	0.06415	-0.37731	0.89722	-0.04381	0.04557	-0.21085	0.88226	0.04471	0.03723	-0.10297
0.3775	0.88788	-0.21485	0.04155	-0.33972	0.88122	0.16841	0.02802	-0.19367	0.86716	0.13489	0.02306	-0.09837
0.4662	0.87346	0.07895	0.02533	-0.30240	0.86834	0.23989	0.02136	-0.16942	0.85424	0.20330	0.01845	-0.08628
0.5839	0.85734	0.15365	0.02023	-0.23644	0.85242	0.25185	0.01792	-0.13186	0.83810	0.23398	0.00907	-0.07351
0.6643	0.84888	-0.10381	0.01748	-0.19065	0.84276	0.13531	0.00842	-0.11334	0.82732	0.26972	0.00711	-0.06033
0.8087	0.83092	-0.10702	0.00764	-0.11333	0.82471	0.10822	0.00660	-0.06485	0.81032	0.07702	0.00583	-0.03443
0.8767	0.82298	-0.14654	0.00674	-0.07317	0.81660	0.07550	0.00555	-0.04221	0.80214	0.05071	0.00497	-0.02240
1.0000	0.80754	0.00000	0.00549	0.00000	0.80242	0.00000	0.00481	0.00000	0.78774	0.00000	0.00425	0.00000
X_1cyclohexanol + $X_2$2-heptanone												
X_2	ρ(g.cm^{-3}) 293.15K	V^E cm^{3}mol^{-1} 293.15K	η 293.15K	η^Emp.s 293.15K	ρ(g.cm^{-3}) 303.15K	V^E cm^{3}mol^{-1} 303.15K	η 303.15K	η^Emp.s 303.15K	ρ(g.cm^{-3}) 313.15K	V^E cm^{3}mol^{-1} 313.15K	η 313.15K	η^Emp.s 313.15K
0.0000	0.94182	0.00000	0.60916	0.00000	0.93940	0.00000	0.35321	0.00000	0.92548	0.00000	0.19250	0.00000
0.0539	0.93282	0.00278	0.26566	-0.31106	0.92562	0.52095	0.15365	-0.18087	0.91501	0.13472	0.09717	-0.08525
0.1596	0.90482	1.39311	0.14947	-0.36363	0.90304	1.19597	0.09716	-0.20071	0.89756	0.14615	0.06390	-0.09878
0.2460	0.89229	1.40108	0.09012	-0.37099	0.88902	1.32880	0.05956	-0.20835	0.88400	0.17353	0.04225	-0.10429
0.3395	0.87840	1.54931	0.05586	-0.34897	0.87506	1.42017	0.04007	-0.19541	0.87014	0.19992	0.02662	-0.10245
0.4364	0.86994	0.97666	0.03740	-0.30911	0.86528	0.96078	0.02509	-0.17679	0.85240	0.83448	0.02114	-0.08984
0.5331	0.86526	-0.08476	0.02405	-0.26427	0.85760	0.26390	0.02075	-0.14760	0.84130	0.62948	0.01783	-0.07508
0.6420	0.85286	-0.18991	0.01978	-0.20299	0.84468	0.18741	0.01740	-0.11319	0.82999	0.31417	0.00830	-0.06428
0.7586	0.83958	-0.18409	0.00990	-0.14270	0.83119	0.17894	0.00847	-0.08168	0.81656	0.30551	0.00718	-0.04361
0.8739	0.82718	-0.17315	0.00827	-0.07494	0.81896	0.11622	0.00720	-0.04297	0.80480	0.16958	0.00611	-0.02314
1.0000	0.81348	0.00000	0.00732	0.00000	0.80664	0.00000	0.00645	0.00000	0.79280	0.00000	0.00570	0.00000

Table (3): Experimental values of density and viscosity and calculated values of excess molar volume and excess viscosity for ternary mixture of X_1cyclohexanol + X_2 2-hexanone + $X_3$2-heptanone at (293.15, 303.15, and 313.15)K.

X_1cyclohexanol + X_2 2-hexanone + X_3 2-heptanone													
X_1	X_2	ρ (g.cm^{-3}) 293.15K	ρ (g.cm^{-3}) 303.15K	ρ (g.cm^{-3}) 313.15K	V^E_{123} cm^{3}mol^{-1} 293.15K	V^E_{123} cm^{3}mol^{-1} 303.15K	V^E_{123} cm^{3}mol^{-1} 313.15K	η 293.15K	η 303.15K	η 313.15K	η^E_{123} mp.s 293.15K	η^E_{123} mp.s 303.15K	η^E_{123} mp.s 313.15K
0.0458	0.4818	0.81638	0.80820	0.79340	-0.13422	0.23357	0.33666	0.65310	0.56574	0.49190	0.61910	0.54419	0.47834
0.1089	0.4495	0.82512	0.81610	0.80146	-0.42433	0.08911	0.15806	0.70960	0.62023	0.54499	0.63756	0.57675	0.51959
0.1705	0.4154	0.83290	0.82319	0.80864	-0.55287	0.07952	0.13270	0.79958	0.67501	0.58222	0.69040	0.61012	0.54526
0.2373	0.3810	0.83866	0.82966	0.81488	-0.25800	0.28595	0.38254	0.87220	0.76328	0.65190	0.72276	0.67517	0.60242
0.2816	0.3566	0.84378	0.83480	0.82012	-0.24374	0.30788	0.38912	0.96190	0.83480	0.70530	0.78576	0.73128	0.64751
0.3312	0.3331	0.84963	0.84006	0.82586	-0.22985	0.42235	0.43319	0.01801	0.90726	0.75979	-0.18803	0.78651	0.69270
0.3956	0.2972	0.85740	0.84740	0.83310	-0.21021	0.51843	0.54961	0.01989	0.01762	0.89974	-0.22498	-0.12552	0.82057
0.4462	0.2707	0.86532	0.85258	0.83880	-0.42755	0.69724	0.65884	0.02163	0.01858	0.97300	-0.25374	-0.14215	0.88434
0.4954	0.2446	0.87199	0.86080	0.84724	-0.47220	0.43661	0.36071	0.02406	0.02048	0.01796	-0.28096	-0.15735	-0.07993
0.5468	0.2180	0.87986	0.86768	0.85416	-0.61828	0.43008	0.35215	0.02710	0.02290	0.01930	-0.30891	-0.17280	-0.08822
0.5966	0.1913	0.88752	0.87464	0.86100	-0.74018	0.40381	0.34594	0.03727	0.02588	0.02152	-0.32876	-0.18713	-0.09534
0.6444	0.1649	0.89498	0.88200	0.86868	-0.84930	0.30767	0.20718	0.04474	0.03669	0.02432	-0.35010	-0.19294	-0.10151
0.6885	0.1437	0.89758	0.88576	0.87306	-0.38602	0.63604	0.46649	0.05780	0.04216	0.02723	-0.36363	-0.20280	-0.10687
0.7433	0.1155	0.90418	0.89418	0.88180	-0.22870	0.57156	0.36562	0.07341	0.04542	0.03827	-0.38104	-0.21859	-0.10611
0.7903	0.0882	0.91099	0.90200	0.88968	-0.22350	0.46006	0.25107	0.09838	0.06458	0.04484	-0.38441	-0.21577	-0.10836
0.8403	0.0642	0.91776	0.90979	0.89804	-0.13676	0.43744	0.16421	0.12922	0.07824	0.05585	-0.38372	-0.21949	-0.10672
0.8848	0.0382	0.92428	0.91686	0.90510	-0.10849	0.41372	0.15031	0.17265	0.09718	0.07385	-0.36711	-0.21602	-0.09707
0.9184	0.0259	0.92916	0.92231	0.91092	-0.06315	0.40534	0.10308	0.22299	0.13189	0.08325	-0.33701	-0.19299	-0.09396

Table (4): Parameter and standard deviations of the Redlich-Kister polynomial equation for binary mixtures studied here at (293.15K, 303.15K, and 313.15K).

T/K		A_0	A_1	A_2	A_3	σ
	X_1 Cyclohexanol + X_2 2-hexanone					
V^E cm^3/mol^{-1}	293.15	15.323	-6.964	2.539	3.682	0.009
	303.15	15.722	-6.874	0.521	-4.872	0.002
	313.15	15.765	-6.738	3.224	-6.045	0.003
η^E mpa.s	293.15	14.444	-7.177	-0.06	-0.791	0.007
	303.15	14.393	-6.765	-0.018	-1.564	0.006
	313.15	14.353	-6.498	-0.005	-0.005	0.004
	X_1 Cyclohexanol + $X_2$2- heptanone					
V^E cm^3/mol^{-1}	293.15	8.853	-7.556	2.41	4.521	0.001
	303.15	10.091	-6.295	1.372	1.079	0.024
	313.15	15.916	-6.109	2.605	-0.918	0.013
η^E mpa.s	293.15	13.824	-6.830	-0.887	1.478	0.008
	303.15	13.745	-6.403	-0.092	-0.001	0.005
	313.15	13.666	-6.117	-0.531	-0.095	0.007

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Figure 1: Excess molar volume V_{12}^{E} Versus X_2 for X_1 cyclohexanol + X_2 hexanone at ■293.15K, ●303.15K and ▲313.15K.

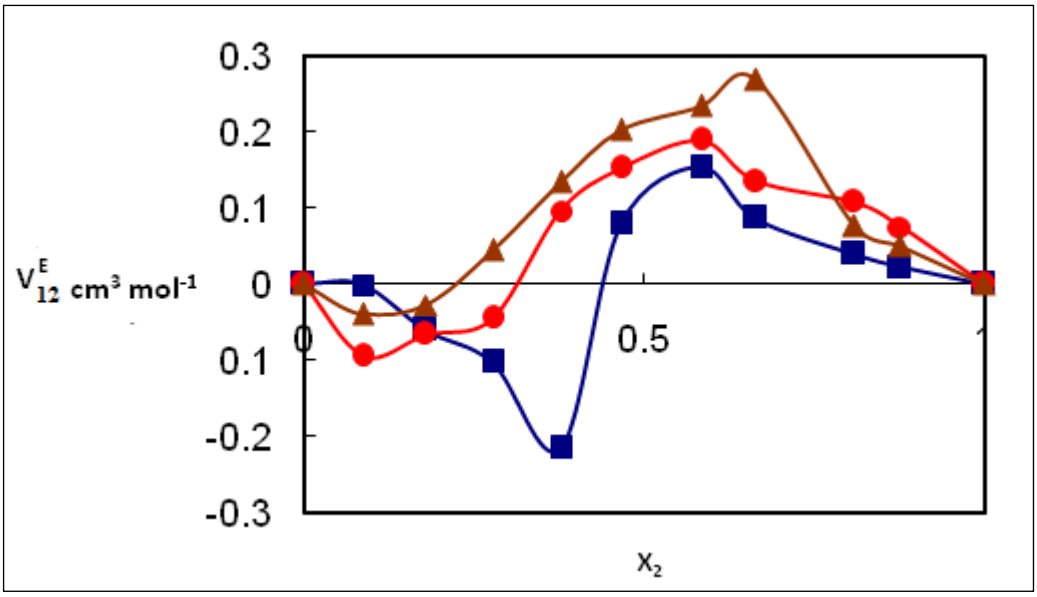


Figure 2: Excess molar volume V_{12}^{E} Versus X_2 for X_1 cyclohexanol +X_2 heptanone at ■293.15K, ●303.15K and ▲313.15K.

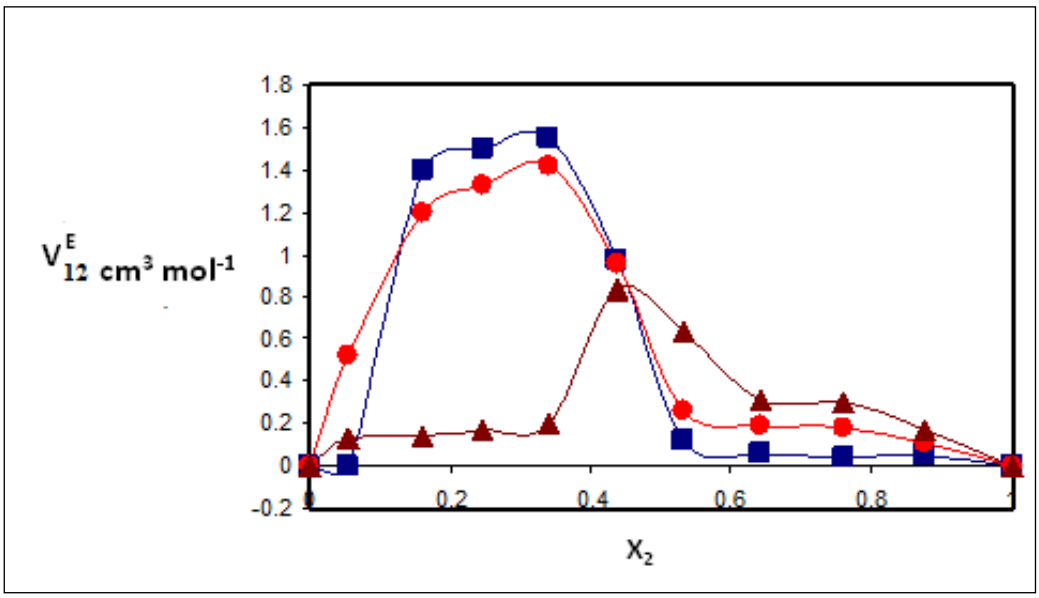
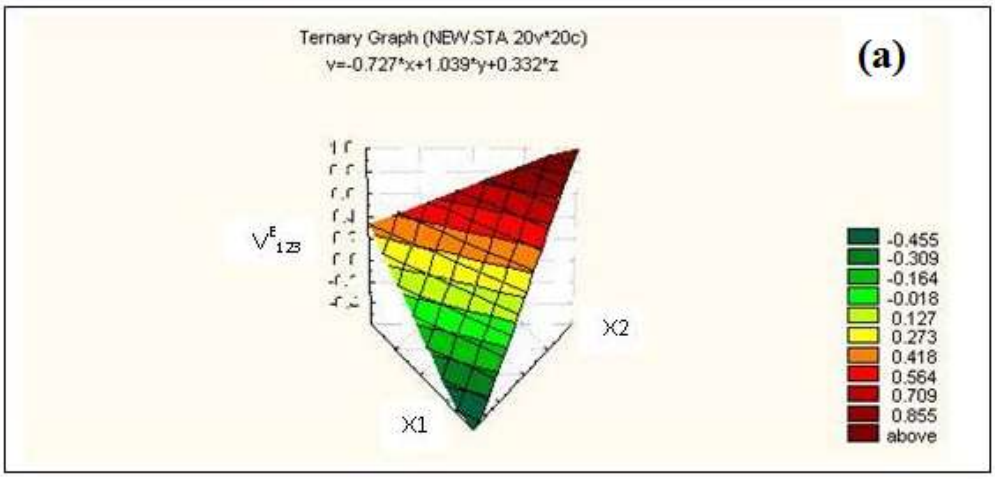
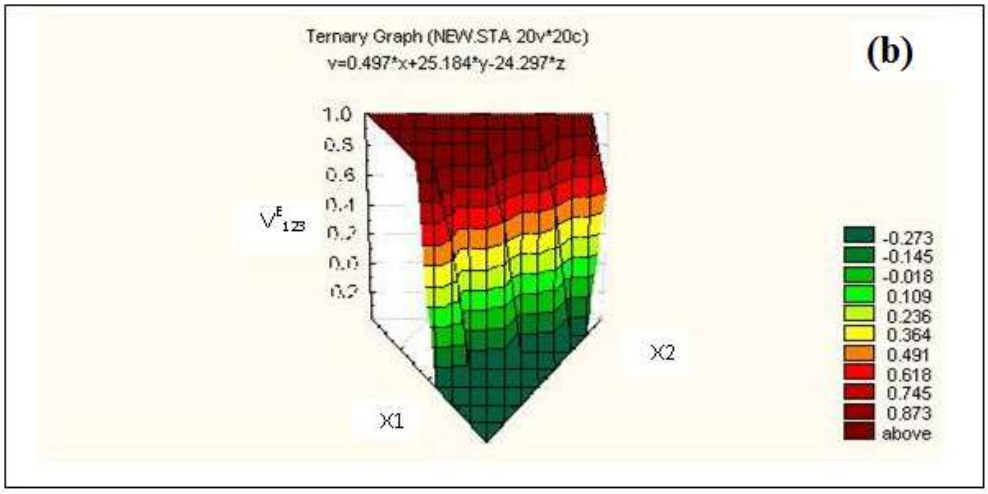


Figure (3): Excess molar volume V_{123}^{E} for ternary system X_1 cyclohexanol + X_2 2-hexanone + X_3 2-heptanone at (a) 293.15K, (b) 303.15K, and (c) 313.15K.





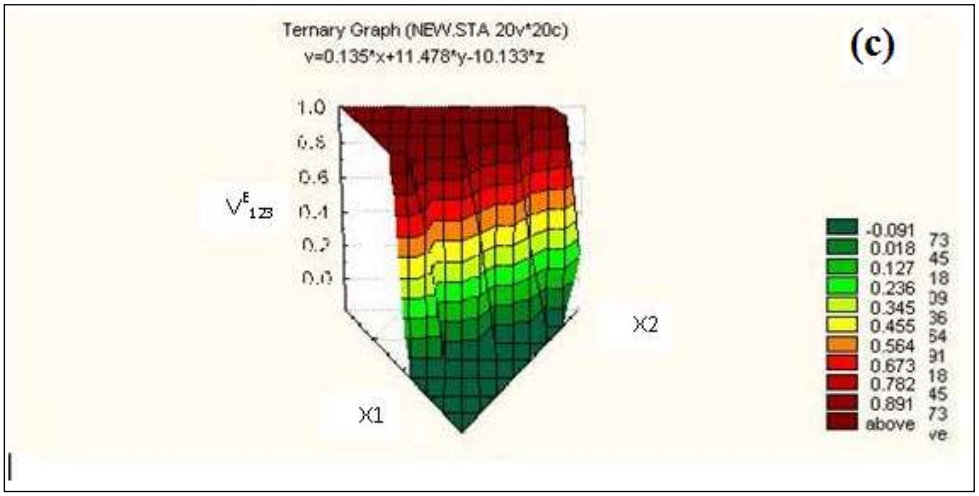


Figure 4: Excess viscosity η_{12}^{E} Versus X_2 for X_1 cyclohexanol +X_2 2- hexanone at ■293.15K, ●303.15K and ▲313.15K.

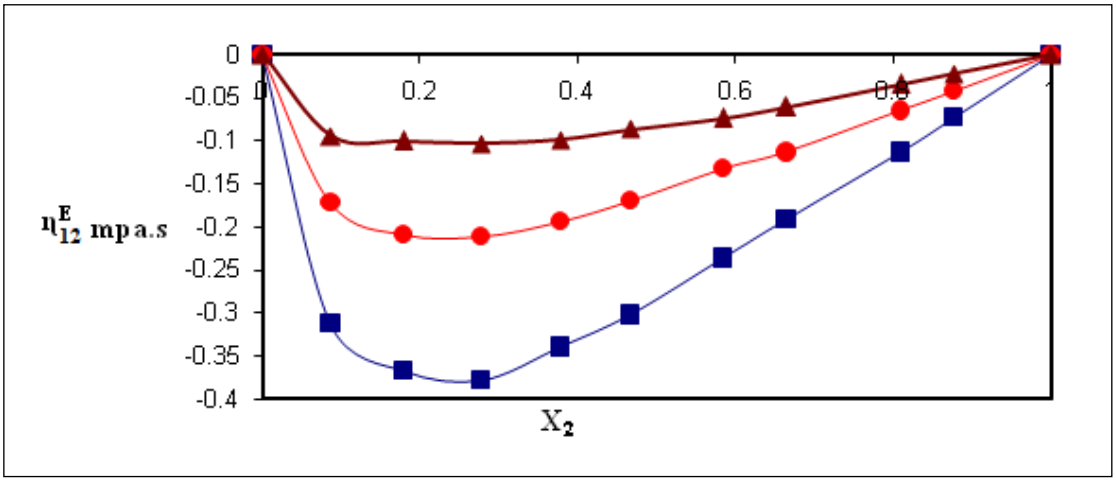


Figure 5: Excess viscosity η_{12}^{E} Versus X_2 for X_1 cyclohexanol +X_2 2- heptanone at ■293.15K, ●303.15K and ▲313.15K.

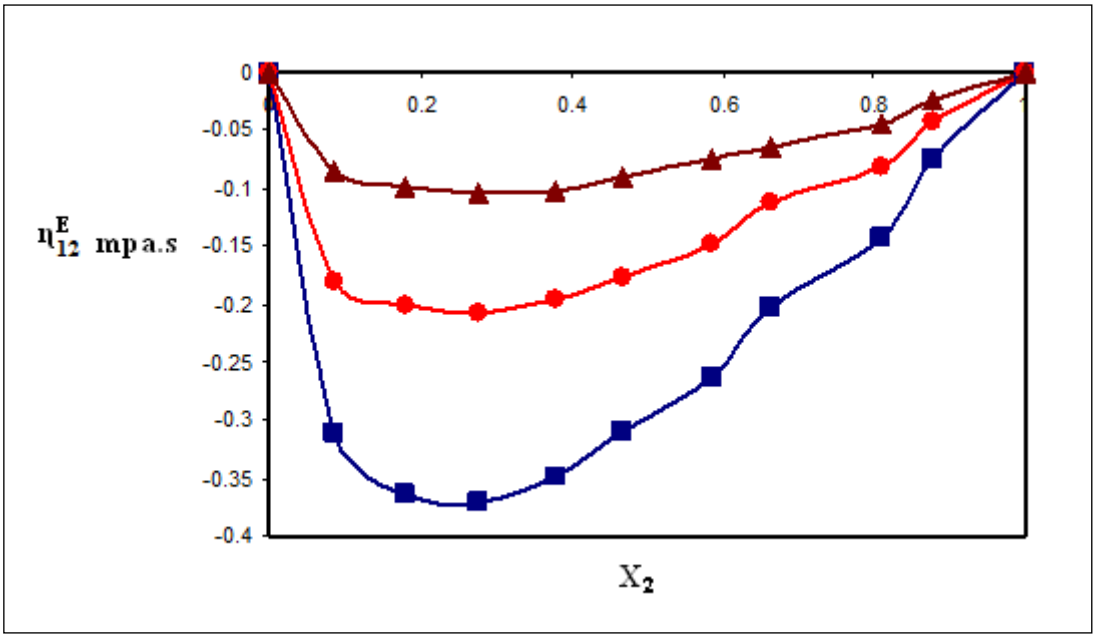


Figure (6): Excess viscosities η^{E}_{123} for ternary system X_1 cyclohexanol + X_2 2-hexanone + X_3 2-heptanone at (a) 293.15K, (b) 303.15K, and (c) 313.15K.