

Study of Molecular interactions in binary mixture of isobutanol with

methyl acetate, ethyl acetate and normal propylacetate at different temperatures

شيماء باسم خلف

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دراسة التداخلات الجزيئية لمزيج ثنائي من آيزوبيوتانول مع كل من خلات المثيل وخلات الاعتيادي في درجات حرارية مختلفة.

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Abstract

The properties associated with liquid mixtures; like density and viscosity,etc.....find several extensive applications, in chemical engineering, design.

These properties are important from practical and theoretical point of view to understand liquid theory and provide informations about molecular interactions.

In this paper, we study the densities and viscosities for binary mixtures of isobutanol with methylacetae , ethylacetate, and n-propylacetate over the whole range of mixture composition at 298.15 , 303.15 and 308.15 K .Excess molar volumes v^E were calculated from precise density measurements at different temperatures. The viscosity deviation η^E where calculated from experimental viscosity values at different temperatures . The observed values of V^E ,and η^E are discussed in term of molecular interactions in these binary mixture. The experimental results were fitted to the Redlich-Kister polynomial equation to evaluate the adjustable parameters and standard deviation for the ideal case .

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ان الخصائص المرتبطة بالسوائل ومزيج السوائل مثل الكثافة و اللزوجة وغير هما لها اهميتها ،حيث تجد هذه الخصائص المرتبطة تطبيقات في تصميم الهندسة الكيميائية. كما ان هذه الخواص مهمة من الناحية النظرية والعملية لفهم نظرية السؤال ودعم المعلومات بشأن التداخلات الجزيئية و قد تم در اسة الكثافة و اللزوجة لمزيج من ايزوبيوتانول مع خلال المثيل وخلات الاثيل وخلات البروبيل الاعتيادي لمدى من الكسور المولية في درجات الحرارة ($^{79.10}$ و $^{79.10}$ و من الكسور المولية في درجات الحرارة المختلفة كما تم حساب الحجم المولي الفائض $^{70.10}$ من قياسات الكثافة ولدرجات الحرارة المختلفة كما تم حساب اللزوجة الفائضة من قياسات اللزوجة ولدرجات الحرارة نفسها و كما تمت مناقشة النتائج وفق التداخلات بين الجزئيات وتكسر وتكوين الاواصر تم تعيير القيم العملية لجميع الدوال الفائضة باستخدام معادلة Redlich – Kister معادلة واستخدام النتائج لحساب الانحراف القياسي عن المثالية لهذه القيم.

Introduction:

The study of thermodynamic and transport properties of pure liquids and liquid mixtures are the subject of scientific research in many laboratories for many years. The thermodynamic excess functions such as molar volume, viscosity, molar enthalpy, molar Gibbs energy and refractive index are a source of information about interaction between molecules of the system components (1)

The systems alkanol + alkanoate are of great interest from a theoretical point of view because in mixing processes the breaking of H–bond structure of the alcohol occurs and the formation of new H–bonded molecular species between the alcohol and the ester takes place. (2.3)

The mixtures of isobutanol are used as a solvent for many purposes like printing inks, solubilizer in textile industry, extraction in the production of drugs antibiotics, hormones, vitamins extractor and as additives in polishes and cleaners ⁽⁴⁾. It is used in the production of many compounds. On the other hand, esters find applications as plasticizes in polymer–processing industries in order to impart favorable thermoplastic behavior. ⁽⁵⁾

An understanding of the mixing behavior of isobutanol with esters is therefore important and has applications in many engineering areas and there was an earlier study on some of thermodynamic properties⁽⁶⁻⁹⁾, we reported here the values of density, viscosity for the binary mixture of isobutanol with methylacetate, ethylacetate and n-propylacetate over the entire range of mixture composition at 298.15, 303.15, and 308.15K.

Experimental

Materials: High purity spectroscopic and analytical grade of methyl acetate and ethylacetate were produced from Aldrich chemical company. isobutanol and propylacetate were provided from E. Merck (Germany). All Samples were used without further purification because their purities exceeded 99% as tested by gas chromatography (HP 6890 series) using a flame ionization detector with a packed colum. Experimental values of ρ and η along with the



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mol% purity of the liquids at 298.15 K are given in table (1). These results agreed well with the published data.

Mixtures were prepared in specially designed glass–stoppered bottles, and the measurements were performed on the same day. An electrical Sartorius balance (precision $1*10^{-5}$ g) was used. The error in mole fraction is around ± 0.0002). (

Table-1-Densities, ρ , and Viscosities, η ,of pure components and those of literature at 298.15K

component	$\rho/\mathrm{g.cm}^{-3}$		η/m.pa.s	
		NAL		
	exp	lit	exp	Lit
Isobutanol	0.7980	$0.7998^{(10)}$	3.350	3.333 ⁽¹⁰⁾
Methylacetate	0.9284	0.9282 ⁽¹¹⁾	0.384	0.385 ⁽¹¹⁾
Ethylacetate	0.8946	0.8945 ⁽¹²⁾	0.424	0.426 ⁽¹²⁾
Propylacetate	0.8830	0.8830 ⁽¹²⁾	0.551	0.553 ⁽¹²⁾

Measurements:

All binary mixture of solvents were prepared by using determined volumes to obtain samples covering the whole range of mole fraction (o > x > 1).

The mixtures were kept in tight stopper bottles to prevent any contamination with air. The mole fraction of each sample (mixture) of solvents was calculated from the following relation:

$$Xi=(Wi/Mi) / \sum_{s=i,j,k}^{m} (Ws/Ms) -----(1)$$

Where W_i and M_i are the weight in gram and the molecular weight of the component liquid. The possible error in the mole fraction is estimated be lower than 0.0001 respectively. Densities of pure liquids and their mixtures were measured by using a digital densimeter (Anton paar DMA 60/602). deionized and degassed water with a specific conductance of $(0.6*10^{-6}~S.cm^{-1})$ was used for calibration . For all mixtures and pure solvents, triplicate measurements were performed and the average is accurate to $\mp 0.0002~\mu g~cm^{-3}$.



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Viscosities were measured by using a Cannon fenske viscometer (sige 75, ASTMD 445 supplied by Industrial Research Glassware Ltd., Rosell, New Jersey). An electrical digital stopwatch with a readability of \pm 0.01s was used for the flow time measurements. The measured viscosity values are accurate to \mp 0.001µpa.s.

Results and Discusion

From the results of densities given in tables(2-4) excess molar volumes have been calculated from the following equation (13)

$$V^{E}=V_{m}-V_{1}X_{1}-V_{2}X_{2}$$
 -----(2)

Where V_m is molar Volume of the mixture, V_1 and V_2 are the molar volumes of components 1 and 2 of the mixture, and X_i represents the mole fraction of the component in the mixture.

From the Values of η of the individual components as well as of the binary mixtures. The $\Delta \eta$ have been calculated from $^{(14)}$

$$\Delta Y = Y_m - Y_1 C_1 - Y_2 C_2 - \dots$$
 (3)

For calculting η^E we have used the mole fractions X_i for C_i

Each set of excess functions have been fitted to the Redlich and Kister equation (15)

$$Y^{E}=X_{1} X_{2} \frac{n}{\sum_{i} A_{j}(X_{2}-X_{i})^{j}}$$
-----(4)

j=o Where Y^E is the excess property, and n is the number of adjustable parameters Aj in each case for all mixtures, the optimum number of adjustable parameters is ascertained from an examination of the variation in standard errors, δ , as given by $^{(16)}$

$$\delta = (Y^{E}) = \left[\frac{(Y_{obs}^{E} - Y_{cal}^{E})^{2}}{N - P} \right]^{1/2} - - - - - - (5)$$

Where N represents the number of experimental points and P is the number of adjustable parameters. The values of these parameters, Aj, along with the standard errors, δ are listed in table (5). Excess molar volume of isobutanol with methylacetate and ethylacetate figures (1-2), are positive over the whole range of mole fraction and become more positive as temperature increase. On the other hand excess molar volume of isobutanol with

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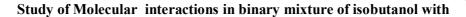
propylacetate are negative figure (3) over the whole range of mole fraction and become more negative as temperature increase.

The observed excess molar volume values in the present investigation may be discussed in terms of several factors which may be divided into physical, chemical and geometrical contributions. The physical interactions involve mainly dispersion forces, giving a positive contribution to $V^{E(17)}$.

In the case of methylacetate the V^E is positive due to the strong intera- molecular hydrogen bond between the methylacetate molecules, so for isobutanol , the interaction between them is very weak . In Ethylacetate the V^E is less positive, which mean that ethylacetate is slowly dissolve in isobutanol because the molecular interaction between the two compounds through the H – bond is stronger than the self – association of them. In n-propylacetate the V^E is negative which mean that interaction between propylacetate and isobutanol molecules is strong due to breaking of intra – H – bond and formation of inter –H – bond with isobutanol (18), the net of isobutanol molecules breaking through the interference of propylacetate molecules and because of it's large molecule will force isobutanol series to expand and give negative V^E . Values of V^E for all binary systems increase with increase of temperature .this is attributed to decreased ester – ester and alkanol – alkanol contacts with an increase in temperature.

Figures (4- 6) shows that the deviations in viscosity are negative in all the systems and become more negative with an increase in chain length of acetate.

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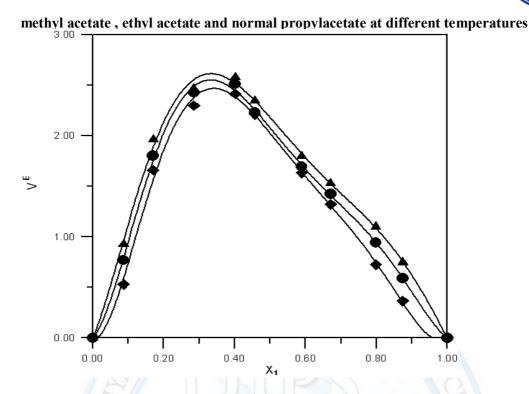


Figure -1-Excess molar volume, V^E , of (X_1 isobutanol + X_2 methylacetate) at different temperatures: •,298.15; • ,303.15, \$\int\$308.15 K.

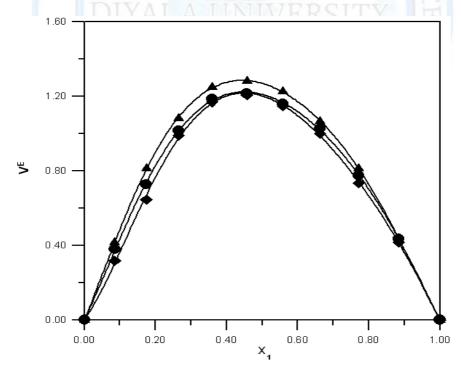


Figure -2-Excess molar volume, V^E , of $(X_1 \text{ isobutanol} + X_2 \text{ ethylacetate})$ at different temperatures: ,298.15; ,303.15, ,303.15 K

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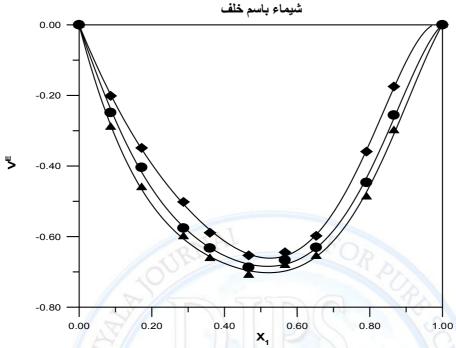
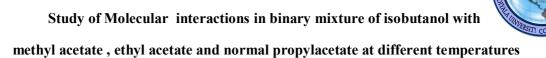


Figure -3-Excess molar volume, V^E , of (X_1 isobutanol + X_2 n- propylacetate) at different temperatures: 4.298.15; 5.298.15, 5.298.15; 5.298.15, 5.298.15;



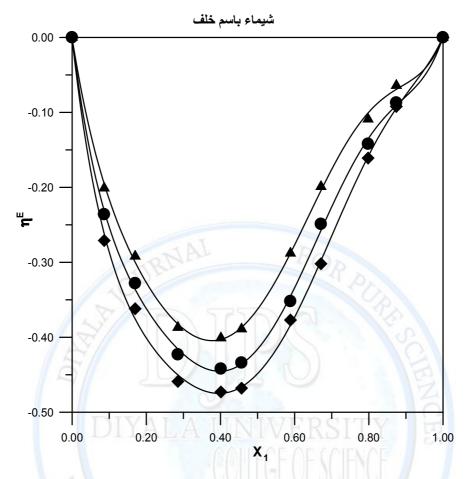


Figure -4-Excess viscosity, η^E ,of (X₁ isobutanol + X₂ methylacetate) at different temperatures: \blacklozenge ,298.15; \bullet ,303.15, \blacktriangle 308.15 K.

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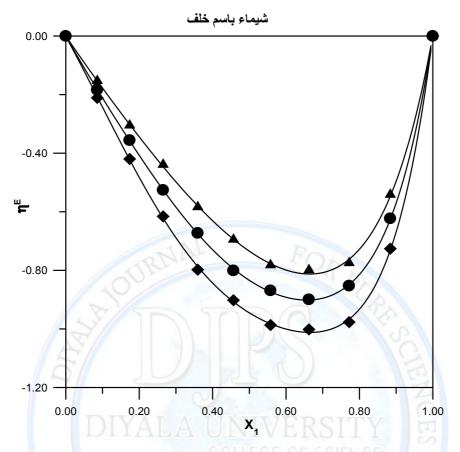
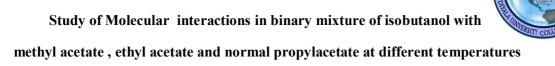


Figure -5-Excess viscosity, η^E ,of (X₁ isobutanol + X₂ ethylacetate) at different temperatures: •,298.15; •,303.15, \triangle 308.15 K.

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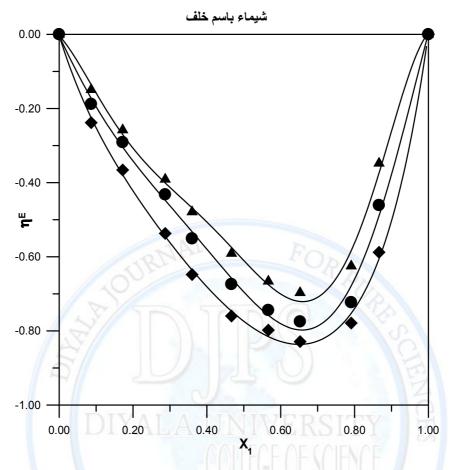


Figure -6-Excess viscosity, η^E ,of (X₁ isobutanol + X₂ n- propylacetate) at different temperatures: •, 298.15; •, 303.15, \clubsuit 308.15 K.

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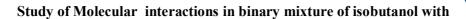
Table -2- Experimental values of density ρ , excess molar volumes V^E , viscosity η and excess viscosity η^E for Binary mixture of X_1 isobutanol $+X_2$ methyl acetate at different temperatures.

X1	$\rho/g.cm^{-3}$	V ^E /cm ³ .mol ⁻¹	η \ср	η ^E \cp
		T=298.15 K		
0.0000	0.9284	0.0000	0.384	0.000
0.0865	0.9095	0.5297	0.485	-0.271
0.1699	0.8853	1.6594	0.620	-0.362
0.2851	0.8633	2.2985	0.900	-0.459
0.4011	0.8472	2.4120	1.130	-0.473
0.4568	0.8422	2.2095	1.527	-0.468
0.5888	0.8313	1.6319	1.783	-0.377
0.6711	0.8243	1.3212	2.202	-0.302
0.7981	0.8147	0.7236	2.685	-0.161
0.8743	0.8091	0.3648	3.001	-0.092
1.0000	0.7980	0	3.350	0.000

T=303.15 K

		P(1) a		
0.00000	09218	0.0000	0.365	0.000
0.0865	0.9006	0.7677	0.459	-0.236
0.1699	0.8781	1.8013	0.568	-0.328
0.2851	0.8576	2.3259	0.807	-0.423
0.4011	0.8413	2.4972	1.005	-0.442
0.4568	0.8371	2.2199	1.360	-0.434
0.5888	0.8261	1.6942	1.589	-0.352

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		,		
0.6711	0.8189	1.4222	1.997	-0.249
0.7981	0.8085	0.9431	2.407	-0.142
0.8743	0.8030	0.5899	2.682	-0.087
1.0000	0.7941	0	2.997	0

T=308.15 K

0.8927 0.8706 0.8508 0.8355 0.8310	0.9257 1.9632 2.4663 2.5791	0.418 0.491 0.669 0.832	-0.201 -0.292 -0.387 -0.401
0.8508 0.8355	2.4663	0.669	-0.387
0.8355			10
7/	2.5791	0.832	-0.401
0.8310			
0.0510	2.3472	1.127	-0.389
0.8205	1.7991	1.348	-0.288
0.8134	1.5284	1.687	-0.199
0.8029	1.1004	2.024	-0.109
0.7975	0.7472	2.249	-0.064
0.7902	0	2.499	0
	0.8134 0.8029 0.7975 0.7902	0.8134 1.5284 0.8029 1.1004 0.7975 0.7472 0.7902 0	0.8134 1.5284 1.687 0.8029 1.1004 2.024 0.7975 0.7472 2.249

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Table -3- Experimental values of density ρ , excess molar volumes V^E , viscosity η and excess viscosity η^E for Binary mixture of X_1 isobutanol $+X_2$ ethylacetate at different temperatures.

X1	ρ/g.cm ⁻³	V ^E /cm ³ .mol ⁻¹	η \ср	η ^E \cp
		T=298.15 K		
0.0000	0.8946	0.0000	0.424	0.000
0.0856	0.8839	0.3174	0.551	-0.211
0.1739	0.8728	0.6433	0.671	-0.420
0.2651	0.8730	0.9881	0.795	-0.616
0.3594	0.8508	1.1674	0.919	-0.798
0.4568	0.8413	1.2056	1.111	-0.902
0.5579	0.8321	1.1467	1.311	-0.987
0.6626	0.8232	0.9981	1.572	-1.002
0.7720	0.8146	0.7324	1.864	-0.977
0.8843	0.8061	0.4144	2.373	-0.726
1.0000	0.7980	0	3.350	0

T=303.15 K

		17 11 0		
0.00000	0.8884	0.0000	0.400	0.000
0.0856	0.8774	0.3783	0.516	-0.184
0.1739	0.8664	0.7258	0.636	-0.356
0.2651	0.8556	1.0153	0.750	-0.526
0.3594	0.8455	1.1841	0.875	-0.673
0.4568	0.8362	1.2162	1.009	-0.801
0.5579	0.8272	1.1601	1.194	-0.869

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0.6626	0.8184	1.0256	1.409	-0.899
0.7720	0.8100	1.7741	1.692	-0.853
0.8843	0.8019	0.4348	2.151	-0.623
1.0000	0.7941	0	2.997	0

T=308.15 K

0.00000	0.8825	0.0000	0.385	0.000
0.0856	0.8714	0.4146	0.476	-0.153
0.1739	0.8602	0.8106	0.561	-0.305
0.2651	0.8498	1.0792	0.659	-0.439
0.3594	0.8399	1.2489	0.735	-0.584
0.4568	0.8308	1.2808	0.838	-0.695
0.5579	0.8220	1.2240	0.957	-0.782
0.6626	0.8136	1.0650	1.139	-0.799
0.7720	0.8054	0.8124	1.356	-0.775
0.8843	0.7978	0.4361	1.777	-0.541
1.0000	0.7902	0	2.499	0

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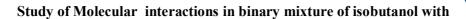
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Table -4- Experimental values of density ρ , excess molar volumes V^E , viscosity η and excess viscosity η^E for Binary mixture of X_1 isobutanol $+X_2$ n-propylacetate at different temperatures.

X1	ρ/g.cm ⁻³	V ^E /cm ³ .mol ⁻¹	η \ср	η ^E \cp
		T=298.15 K		
0.0000	0.8830	0.0000	0.551	0.000
0.0866	0.8785	-0.2013	0.686	-0.238
0.1719	0.8735	-0.3487	0.844	-0.291
0.2870	0.8662	-0.5018	0.986	-0.537
0.3599	0.8612	-0.5891	1.113	-0.648
0.4668	0.8532	-0.6533	1.283	-0.760
0.5676	0.8446	-0.6445	1.544	-0.798
0.6526	0.8368	-0.5980	1.771	-0.775
0.7911	0.8220	-0.3591	2.089	-0.779
0.8664	0.8131	-0.1746	2.519	-0.588
1.0000	0.7980	0	3.350	0

T=303.15 K

0.00000	0.8779	0.0000	0.512	0.000
0.0866	0.8734	-0.2488	0.655	-0.188
0.1719	0.8686	-0.4040	0.773	-0.258
0.2870	0.8615	-0.5761	0.943	-0.432
0.3599	0.8565	-0.6327	1.035	-0.551
0.4668	0.8485	-0.6872	1.163	-0.674



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		,		
0.5676	0.8401	-0.6663	1.358	-0.744
0.6526	0.8327	-0.6559	1.586	-0.697
0.7911	0.8184	-0.4469	1.846	-0.723
0.8664	0.8096	-0.2559	2.320	-0.461
1.0000	0.7941	0	2.997	0

T=308.15 K

0.0000	0.8718	0.0000	0.483	0.000	
0.0866	0.8682	-0.2901	0.603		
0.1719	0.8628	-0.3450	0.646	-0.258	
0.2870	0.8565	-0.6002	0.792	-0.391	
0.3599	0.8516	-0.6621	0.875	-0.479	
0.4668	0.8438	-0.7096	0.966	-0.591	
0.5676	0.8354	-0.6817	1.107	-0.666	
0.6526	0.8317	-1.1010	1.223	-0.697	
0.7911	0.8144	-0.4877	1.527	-0.625	
0.8664	0.8058	-0.2999	1.976	-0.348	
1.0000	0.7902	PROVE 0	2.499	0	

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Study of Molecular interactions in binary mixture of isobutanol with

methyl acetate, ethyl acetate and normal propylacetate at different temperatures

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Table (5) - Redlich-Kister coefficients A_i and standard deviations, σ , for the excess molar quantities of solution of $(x_1 \text{ isobutanol+} x_2 \text{ acetates mixtures})$ at different temperatures.

System	property	T/K	\mathbf{a}_0	a ₁	$\mathbf{a_2}$	a ₃	σ
IsobutanoI+methylacetate	V ^E /cm ³ .mol ⁻¹	298.15	-0.491	0.050	-0.149	0.491	0.003
		303.15	-0.528	0.027	-1.171	0.660	0.004
		308.15	-0.580	-0.109	-0.092	1.175	0.011
	Δη/mPa.s	298.15	-0.176	0.160	-0.131	0.034	0.001
		303.15	-0.191	0.120	-0.122	0.217	0.001
		308.15	-0.204	0.068	-0.107	0.453	0.003
Isobutanol+ethylacetate	DIYALA	298. 15	1.169	0.412	-0.228	-0.367	0.005
	V ^E /cm ³ .mol ⁻¹ Δη/mPa.s	303. 15	1.295	0.496	-0.153	-0.544	0.004
		308. 15	1.405	0.594	0.006	-0.683	0.005
		298. 15	-0.967	0.459	-0.209	0.349	0.005
		303. 15	-0.882	0.537	-0.249	0.085	0.005
		308. 15	-0.882	0.537	-0.249	0.085	0.003
	V ^E /cm ³ .mol ⁻¹	298. 15	0.757	-0.460	-0.148	0.320	0.004
Isobutanol+propylacetate		303. 15	0.962	-0.488	-0.125	0.209	0.005

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	,					
	308.	1.107	-0.420	0.184	0.237	0.004
	15	1.107	-0.420	0.104		
	298.	-2.279	1.625	-1.015	0.316	0.002
	15					
Δη/mPa	_{.s} 303.	303. 15	1.443	-0.789	0.105	0.008
					00100	
	308.	308. 15	711 1.276	-0.686	-0.094	0.010
	15					0.010

