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VOLUMETRIC AND VISCOMETRIC PROPERTIES OF BINARY LIQUID MIXTURES OF ACRYLIC ESTERS WITH HEPTANE-2-OL AT 298.15 AND 308.15 K TEMPERATURES

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ABSTRACT

New experimental thermodynamic data involving density and viscosity of the binary liquid mixture methyl acrylate, ethyl acrylate and butyl acrylate with heptane-2-ol have been measured over entire range of composition, at 298.15 and 308.15 K temperatures and at atmospheric pressure. These basic parameters were correlated by recently proposed Jouyban-Acree model. Density and viscosity were also used to evaluate excess molar volume and viscosity deviation respectively; their values were fitted in Redlich-Kister polynomial equation. The mixture viscosities were correlated using Grunberg-Nissan, Tamura and Kurata, and McAllister three and four body model equations.

Keywords: Acrylic esters, heptane-2-ol, Jouyban Acree model, Redlich-Kister Equation, McAllister model.

INTRODUCTION

Alcohols serve as simple example of biologically and industrially important amphiphilic materials [1]. These exists as associated structures in liquid state and this association may be due to hydrogen bonding of their - OH group. In higher alcohols geometrical fitting of one into other is negligibly small; hence association decreases with increase in chain length of alcohols. Esters of both aliphatic and acrylic types are important industrial chemicals. Acrylic esters are unique molecule with unsaturated structures along side a carbonyl group [2]. Thermodynamic studies of binary systems are also useful in solving many problems associated with heat and fluid flow. The qualitative and quantitative analysis of excess functions provides information about the nature of molecular interactions in the binary mixtures. Thermodynamic properties are also important in designing industrial equipments with better precision. Excess thermodynamic functions are dependent not only on difference in intermolecular forces but also on difference in size and shape of molecules. Literature survey reveals that most of work has been done on acrylic esters with alkane-1-ols [3-9].

EXPERIMENTAL

Chemicals used in the present study were of analytical grade and supplied by S.D. Fine Chemicals Pvt., Mumbai (India). Prior to use all liquids were stored over 0.4 nm molecular sieves to reduce the water content and were degassed. In addition, all three acrylic esters were distilled before use. The masses were recorded on a Mettlar one pan balance, which can read up to fifth place of decimal , with an accuracy of ± 0.01 mg. Care was taken to avoid evaporation and contamination during mixing. The estimated uncertainty in mole fraction was $<1\times10^{-4}$. The densities[10] of the solutions were measured using a single capillary pycnometer made up of borosil glass with a bulb of $8 cm^3$ and capillary with internal diameter of 0.1cm. The reproducibility of density measurement was $\pm 5x10^{-5}$ g/cm^3.The dynamic viscosities were measured using an Ubbelohde suspended level viscometer[10] calibrated with conductivity water. An electronic digital stop watch with readability of ± 0.01 s was used for the flow time measurements. Since all flow times were greater than 300 s and capillary radius (0.1 mm) was far less than its length (50 to 60) mm, the kinetic energy and end corrections, respectively, were negligible. The uncertainty in dynamic viscosities is $\pm 3 x10^{-3}$ mPa.s.

RESULTS AND DISCUSSION

The excess molar volumes, V^E were calculated using following equation,

$$V^E / \text{cm}^3\text{mol}^{-1} = [x_1M_1+x_2M_2]/\rho_{12} - [\,(x_1M_1/\rho_1)+(\,x_2M_2/\rho_2)] \tag{1}$$

where ρ_{12} is the density of the mixture and x_1, M_1, ρ_1, and x_2, M_2, ρ_2 are the mole fraction, the molecular weight, and the density of pure components 1and 2, respectively. The viscosity deviations ($\Delta\eta$) were calculated using equation,

$$\Delta\eta \,/\text{mPa.s} = \eta_{12} - x_1\eta_1 - x_2\eta_2 \tag{2}$$

where η_{12} is the viscosity of the mixture and x_1, x_2 and η_1, η_2 are the mole fraction and the viscosity of pure components 1and 2 respectively[11]. The density, viscosity, excess molar volume and deviation in viscosity for binary liquid mixture of acrylic ester with heptane-2-ol are listed in Table 2.

Table-1: Densities, ρ, Viscosities, η, for Pure Components at T= (298.15 and 308.15)K.

	T=298.15K		T=308.15K	
	Expt.	Lit.	Expt	Lit.
	Heptane-2-ol			
ρ / (g.m^{-3})	0.81376	0.81374[16]	0.80487	0.80488[16]
η / (mPa.s)	5.085	---	3.382	---
	Methyl Acrylate			
ρ / (g.m^{-3})	0.94751	0.94750[17]	0.93561	0.93770[18]
η / (mPa.s)	0.449	0.449[17]	0.390	0.384[19]
	Ethyl Acrylate			
ρ / (g.m^{-3})	0.91632	0.91593[18]	0.90400	0.90460[19]
η / (mPa.s)	0.518	0.530[18]	0.456	0.455[17]
	Butyl Acrylate			
ρ / (g.m^{-3})	0.89399	0.89395[18]	0.88546	0.88460[19]
η / (mPa.s)	0.787	---	0.684	0.684[18]

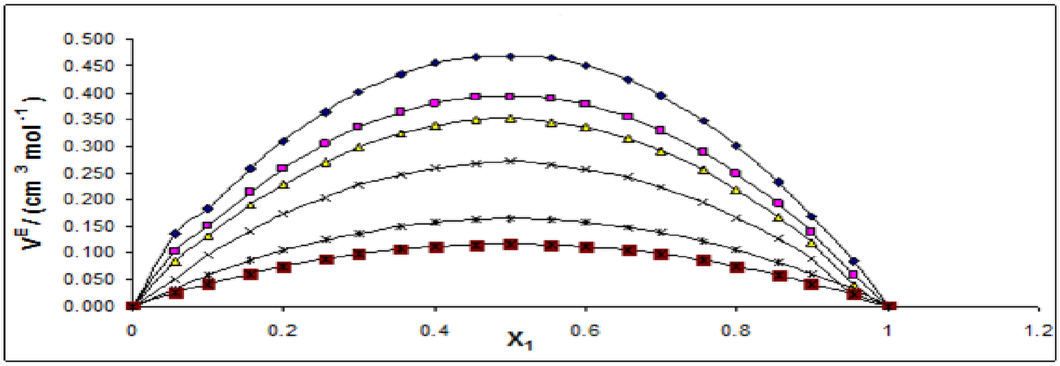

Fig.-1: Variation of excess molar volumes for binary mixtures of Acrylic esters (1) + Heptane-2-ol (2) at 298.15 K:♦, Methyl acrylate; ▲,Ethyl acrylate; *, Butyl acrylate and at 308.15 K: ▪, Methyl acrylate; ×,Ethyl acrylate; ■, Butyl acrylate

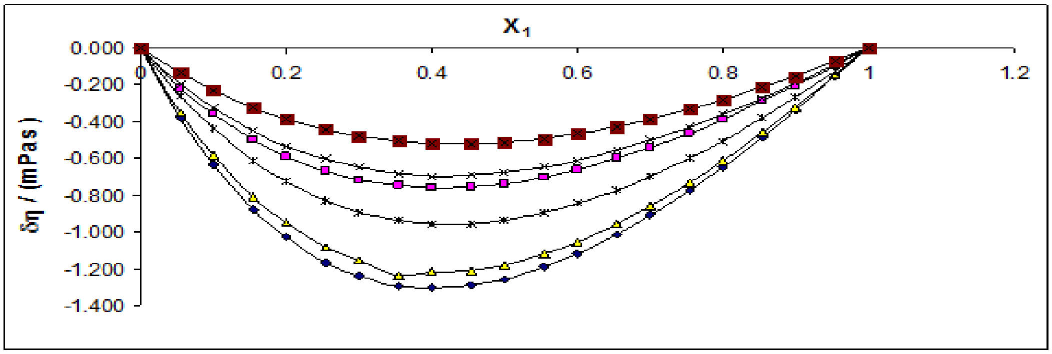

Fig.-2: Variation of deviation in viscosity for binary mixtures of Acrylic esters (1) + Heptane-2-ol (2) at 298.15 K: ♦, Methyl acrylate; ▲,Ethyl acrylate; ∗, Butyl acrylate and at 308.15 K: ■, Methyl acrylate; ×,Ethyl acrylate; ■,Butyl acrylate

The excess molar volumes and deviations in viscosity were fitted to Redlich- Kister equation of the type,

$$Y = x_1x_2 \sum_{i}^{n} a_i (x_1 - x_2)^i \qquad (3)$$

Where Y is either V^E or $\Delta\eta$ or κ_s^E and n is the degree of polynomial. Coefficient a_i was obtained by fitting Eq 3 to experimental results using a least-squares regression method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (σ).

σ was calculated using the relation,

$$\sigma\,(Y) = \left[\frac{\sum (Y_{\exp t} - Y_{calc})^2}{N-n}\right]^{1/2} \qquad (4)$$

Where N is the number of data points and n is the number of coefficients [12]. The calculated values of the coefficients a_i with standard deviations (σ) are given in Table 3.

Several relations like Grunberg-Nissan, Tamura and Kurata having one adjustable parameter have been proposed to evaluate the dynamic viscosity η of liquid mixtures. Grunberg-Nissan provided the following empirical equation,

$$\ln \eta_{12} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2G_{12} \qquad (5)$$

where G_{12} is a parameter proportional to the interchange energy.

Tamura and Kurata developed expression for viscosity of binary mixtures as,

$$\eta = x_1\phi_1 \eta_1 + x_2\phi_2 \eta_2 + 2(x_1x_2\phi_1\phi_2)^{1/2}T_{12} \qquad (6)$$

where T_{12} is the interaction parameter , ϕ_1 and ϕ_2 are the volume fractions.

McAllister's multibody interaction model was widely used to correlate kinematic viscosity (υ) data. The two parameter McAllister equation based on Eyring's theory of absolute reaction rates, taken into account interactions of both like and unlike molecules by a two dimensional three body model. The three body model given by relation,

$$\ln v = x_1^3 \ln v_1 + x_2^3 \ln v_2 + 3x_1^2 x_2 \ln Z_{12} + 3x_1 x_2^2 \ln Z_{21} - \ln [x_1 + (x_2M_2/M_1)] + 3x_1^2 x_2 \ln[(2/3) + (M_2/3M_1)] + 3x_1x_2^2 \ln[(1/3) + (2M_2/3M_1)] + x_2^3 \ln(M_2/M_1) \qquad (7)$$

Similarly, the four body model was defined by the relation,

$$\ln v = x_1^4 \ln v_1 + 4x_1^3x_2 \ln Z_{1112} + 6x_1^2x_2^2 \ln Z_{1122} + 4x_1x_2^3 \ln Z_{2221} + x_2^4 \ln v_2 - \ln [x_1 + x_2(M_2/M_1)] + 4x_1^3x_2 \ln [(3 + M_2/M_1)/4] + 6x_1^2\, 2x_2^2 \ln[1 + M_2/M_1)/2] + 4x_1x_2^3 \ln [(1 + 3M_2/M_1)/4] + x_2^4 \ln (M_2/M_1) \qquad (8)$$

Where Z_{12}, Z_{21}, Z_{1112}, Z_{1122} *and* Z_{2221} are model parameters and M_i and v_i are the molecular mass and kinematic viscosity of pure component *i*.

To perform a numerical comparison of the correlating capability of above Eq 5 to 8 we have calculated the standard percentage deviation (σ %) using the relation,

$$\sigma\ \% \ = [1/ (\eta_{expt} - \mathrm{k}) \times \sum (100 (\eta_{expt} - \eta_{cal}) / \eta_{expt})^2]^{1/2} \qquad (9)$$

where k represents the number of numerical coefficients in the respective equations.

The terms G_{12}, T_{12}, Z_{12}, Z_{21}, Z_{1112}, Z_{1122} *and* Z_{2221} were estimated by a non-linear regression analysis based on a least-squares method [13]. These are presented with their standard percentage deviation (σ %) in Table 4.

Recently Jouyban and Acree proposed a model for correlating the density and viscosity of liquid mixtures at various temperatures. The proposed equation is,

$$\ln y_{mT} = f_1 \ln y_{1T} + f_2 \ln y_{2T} + f_1 f_2 \sum [A_j (f_1 - f_2)^j / T] \qquad (10)$$

where y_{mT}, y_{1T} and y_{2T} is density or viscosity of the mixture and solvents 1 and 2 at temperature *T*, respectively, f_1 and f_2 are the volume fractions of solvents in case of density, and mole fraction in case of viscosity, and *Aj* are the model constants. The correlating ability of the Jouyban - Acree model was tested by calculating the average percentage deviation (APD) between the experimental and calculated density and viscosity as,

$$APD = (100/N) \sum [(|\, y_{expt} - y_{cal} \,|)/ y_{expt})] \qquad (11)$$

Where *N* is the number of data points in each set. The optimum numbers of constants *Aj,* in each case, were determined from the examination of the average percentage deviation value. Constants *Aj* calculated from the least square analysis with the average percentage deviation (APD) are presented in Table 5. This model can be used in data modeling [14, 15].

A graphical comparison of the dependence of excess molar volume, V^E, at 298.15 and 308.15K for the binary mixtures of each acrylic ester with heptane-2-ol is given in Fig. 1.Excess molar volumes found to be positive and increases with increase in chain length of acrylic esters molecules and with temperature due to decrease of dipolar association of the components and dissociation of self-associated compounds. The magnitude and sign of V^E is a reflection of the type of interactions taking place in the mixture. The overall positive magnitude of indicates a net dislocation effect and is caused by an intercalation effect of heptane-2-ol that breaks dipole-dipole associations. The disruption of alkane-2-ol multimer through breaking of hydrogen bond makes a positive contribution to V^E . Non specific physical interactions between real species present in mixture also contribute positive deviation to V^E . A graphical comparison of the dependence of deviation in viscosity, $\Delta\eta$, at 298.15 and 308.15K for the binary mixtures of each acrylic ester with heptane-2-ol is given in Fig. 2. Decrease in viscosity attributed to breaking of dipolar association of alkane-2-ol into small dipoles. Weak types of dipole – induced dipole type interactions are not sufficient to produce bulky or less mobile entities in system and hence decreased trend of viscosity. The main effect in viscosity deviation of mixture is breaking of self interactions in compounds during mixing process.

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Table-2: Densities, ρ, Viscosities, η, Excess Molar Volumes, V^E, and Viscosity Deviation, $\Delta\eta$, for Acrylic esters (1) + Heptane-2-ol (2) at *T*= (298.15 and 308.15) K.

	T=298.15K				T=308.15K			
x_1	ρ/ (g.cm^{-3})	η/ (mPa.s)	V^E/ (cm^{3}mol^{-1})	Δη/ (mPa.s)	ρ/ (g.cm^{-3})	η/ (mPa.s)	V^E/ (cm^{3}mol^{-1})	Δη/ (mPa.s)
MA (1) + Heptane-2-ol (2)								

0	0.81376	5.085	0.000	0.000	0.80487	3.382	0.000	0.000
0.05534	0.81777	4.446	0.137	-0.383	0.80899	3.001	0.103	-0.216
0.09992	0.82149	3.990	0.183	-0.632	0.81263	2.725	0.150	-0.358
0.15545	0.82620	3.487	0.258	-0.878	0.81731	2.417	0.214	-0.500
0.19991	0.83016	3.130	0.310	-1.028	0.82125	2.196	0.258	-0.588
0.25536	0.83536	2.736	0.365	-1.165	0.82640	1.948	0.305	-0.670
0.29977	0.83975	2.456	0.401	-1.239	0.83073	1.770	0.335	-0.715
0.35538	0.84554	2.146	0.435	-1.291	0.83644	1.569	0.364	-0.749
0.39998	0.85042	1.926	0.455	-1.305	0.84126	1.425	0.381	-0.760
0.45505	0.85680	1.685	0.468	-1.290	0.84753	1.265	0.392	-0.755
0.49982	0.86231	1.511	0.466	-1.256	0.85290	1.149	0.393	-0.738
0.55554	0.86946	1.320	0.465	-1.189	0.85994	1.018	0.389	-0.701
0.59984	0.87552	1.186	0.451	-1.119	0.86585	0.925	0.378	-0.662
0.65549	0.88356	1.036	0.424	-1.010	0.87370	0.821	0.355	-0.600
0.69989	0.89037	0.930	0.394	-0.910	0.88034	0.746	0.329	-0.542
0.75499	0.89932	0.814	0.347	-0.771	0.88905	0.662	0.289	-0.461
0.79992	0.90708	0.729	0.300	-0.647	0.89658	0.601	0.250	-0.388
0.85522	0.91723	0.638	0.233	-0.482	0.90642	0.533	0.192	-0.290
0.89985	0.92596	0.572	0.170	-0.341	0.91487	0.484	0.139	0.206
0.95546	0.93752	0.500	0.085	-0.155	0.92612	0.429	0.059	-0.099
1	0.94751	0.449	0.000	0.000	0.93561	0.390	0.000	0.000
EA (1) + Heptane-2-ol (2)								
0	0.81376	5.085	0.000	0.000	0.80487	3.382	0.000	0.000
0.05551	0.81769	4.479	0.084	-0.352	0.80885	3.026	0.052	-0.194
0.09981	0.82101	4.048	0.131	-0.581	0.81209	2.769	0.096	-0.321
0.15536	0.82528	3.566	0.189	-0.810	0.81630	2.477	0.141	-0.450
0.19997	0.82884	3.220	0.228	-0.951	0.81979	2.265	0.173	-0.532
0.25549	0.83341	2.837	0.270	-1.082	0.82428	2.020	0.205	-0.608
0.29991	0.83720	2.563	0.298	-1.152	0.82799	1.854	0.227	-0.650
0.35555	0.84212	2.257	0.324	-1.240	0.83280	1.658	0.247	-0.683
0.39996	0.84618	2.039	0.339	-1.219	0.83675	1.517	0.259	-0.694
0.45543	0.85145	1.797	0.349	-1.209	0.84187	1.358	0.267	-0.692
0.49997	0.85582	1.623	0.352	-1.179	0.84610	1.242	0.272	-0.677
0.55522	0.86146	1.430	0.346	-1.119	0.85157	1.112	0.265	-0.646
0.59982	0.86618	1.292	0.336	-1.054	0.85612	1.017	0.257	-0.610
0.65499	0.87223	1.139	0.315	-0.955	0.86195	0.910	0.241	-0.555
0.69998	0.87735	1.028	0.292	-0.861	0.86687	0.832	0.223	-0.502
0.75498	0.88386	0.906	0.255	-0.731	0.87310	0.745	0.195	-0.428
0.79991	0.88937	0.818	0.219	-0.614	0.87838	0.681	0.167	-0.361
0.85505	0.89641	0.721	0.167	-0.459	0.88509	0.609	0.126	-0.271
0.89985	0.90234	0.651	0.119	-0.324	0.89075	0.557	0.089	-0.192
0.95540	0.91010	0.573	0.039	-0.148	0.89814	0.498	0.022	-0.088
1	0.91632	0.518	0.000	0.000	0.90400	0.456	0.000	0.000
BA (1) + Heptane-2-ol (2)								
0	0.81376	5.085	0.000	0.000	0.80487	3.382	0.000	0.000
0.05544	0.81804	4.585	0.033	-0.262	0.80921	3.095	0.025	-0.137
0.09984	0.82147	4.221	0.058	-0.435	0.81270	2.883	0.042	-0.230
0.15549	0.82578	3.804	0.086	-0.612	0.81708	2.638	0.062	-0.325
0.19986	0.82924	3.502	0.105	-0.724	0.82058	2.457	0.075	-0.386
0.25513	0.83357	3.159	0.124	-0.830	0.82496	2.249	0.089	-0.444

0.29905	0.83702	2.910	0.137	-0.890	0.82845	2.097	0.098	-0.478
0.35543	0.84147	2.620	0.150	-0.938	0.83295	1.916	0.107	-0.507
0.39986	0.84499	2.411	0.157	-0.955	0.83649	1.785	0.113	-0.518
0.45520	0.84939	2.175	0.163	-0.954	0.84093	1.634	0.116	-0.520
0.49991	0.85297	2.000	0.164	-0.936	0.84453	1.521	0.117	-0.512
0.55554	0.85744	1.803	0.162	-0.894	0.84901	1.391	0.116	-0.492
0.59991	0.86102	1.660	0.158	-0.847	0.85260	1.296	0.113	-0.467
0.65520	0.86550	1.497	0.149	-0.772	0.85709	1.187	0.106	-0.428
0.69981	0.86913	1.378	0.139	-0.700	0.86072	1.105	0.099	-0.389
0.75555	0.87369	1.241	0.122	0.596	0.86528	1.011	0.087	-0.333
0.79997	0.87734	1.143	0.106	-0.504	0.86892	0.941	0.076	-0.282
0.85550	0.88193	1.030	0.083	-0.378	0.87348	0.861	0.059	-0.212
0.89991	0.88561	0.948	0.061	-0.269	0.87715	0.802	0.043	-0.152
0.95540	0.89023	0.855	0.032	-0.124	0.88173	0.734	0.023	-0.070
1	0.89939	0.787	0.000	0.000	0.88546	0.684	0.000	0.000

Table-3: Adjustable parameters of Eq 3 and 4 for the Mathematical Representation of Excess Functions for binary liquid mixture of Acrylic esters (1) + Heptane-2-ol (2) at *T*= (298.15 and 308.15) K.

		T/K	a_0	a_1	a_2	a_3	a_4	σ
MA (1) + Heptane-2-ol (2)								
$V^E/(cm^3mol^{-1})$		298.15	1.4813	0.3318	0.7304	-2.8201	-2.5563	0.02906
		308.15	1.5823	0.0957	-0.0701	-0.4267	0.2174	0.00530
Δη/(mPa.s)		298.15	-5.0251	1.9392	-0.5722	0.1331	-0.0315	0.00026
		308.15	-2.9517	1.0263	-0.2624	0.0506	-0.0256	0.00030
EA (1) + Heptane-2-ol (2)								
$V^E/(cm^3mol^{-1})$		298.15	1.3971	0.0970	0.1506	-0.4356	-0.3223	0.00578
		308.15	1.0551	0.0675	0.3584	-0.2911	-0.7963	0.00598
Δη/(mPa.s)		298.15	-4.7156	1.7194	-0.4715	0.1128	-0.0420	0.00029
		308.15	-2.7088	0.8806	-0.2169	0.0300	-0.0127	0.00027
BA (1) + Heptane-2-ol (2)								
$V^E/(cm^3mol^{-1})$		298.15	0.6646	-0.0246	-0.0744	0.1157	0.1325	0.00237
		308.15	0.4797	-0.0240	-0.1074	0.0902	0.2029	0.00241
Δη/(mPa.s)		298.15	-3.7420	1.1293	-0.2666	0.0503	-0.0043	0.00057
		308.15	-2.0481	0.5342	-0.1085	0.0129	-0.0086	0.00047

Table-4: Adjustable parameters of Eq 5,6,7,8 and 9 for binary liquid mixture of Acrylic esters (1) + Heptane-2-ol (2) at *T*= (298.15 and 308.15) K

T/K	G_{12}	σ	T_{12}	σ	Z_{12}	Z_{21}	σ	Z_{1112}	Z_{1122}	Z_{2221}	σ
MA (1) + Heptane-2-ol (2)											
298.15	-0.001	0.034	-0.299	21.363	1.152	2.717	0.094	0.919	1.516	3.332	0.796
308.15	-0.001	0.034	0.068	14.440	0.930	1.991	0.047	0.759	1.159	2.396	1.664
EA (1) + Heptane-2-ol (2)											

298.15	-0.001	0.023	0.101	14.441	1.275	2.840	0.035	1.039	1.767	3.454	1.194
308.15	-0.001	0.041	0.358	9.253	1.033	2.098	0.065	0.862	1.370	2.489	1.108
BA (1) + Heptane-2-ol (2)											
298.15	-0.001	0.030	1.043	4.963	1.691	3.253	0.031	1.436	2.463	3.828	2.463
308.15	-0.001	0.041	1.000	2.916	1.356	2.391	0.047	1.176	1.895	2.749	1.854

Table-5: Adjustable parameters of Eq 10 and 11 for binary liquid mixture of Acrylic esters (1) + Heptane-2-ol (2) at T= (298.15 and 308.15) K.

	a_0	a_1	a_2	a_3	a_4	σ	APD
MA (1) + Heptane-2-ol (2)							
ρ / (g.m^{-3})	-21.0333	-4.5468	-1.2836	2.7280	1.6944	4.9461	0.0286
η / (mPa.s)	-0.5137	1.4207	5.2344	-4.5237	-9.9763	1.8944	0.0262
EA (1) + Heptane-2-ol (2)							
ρ / (g.m^{-3})	-10.0776	-1.3335	-0.8306	0.7926	1.5417	2.8032	0.0207
η / (mPa.s)	-0.3056	0.6470	1.7390	-3.2838	-4.8876	1.9652	0.0226
BA (1) + Heptane-2-ol (2)							
ρ / (g.m^{-3})	0.2965	1.1537	-0.3761	-1.7451	0.6281	0.8281	0.0270
η / (mPa.s)	0.0478	-0.1256	-1.4952	-0.6498	0.5697	2.1714	0.0215

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