

Correlation Analysis of the viscosity Arrhenius-type equations parameters for some binary liquids mixtures

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Abstract: The analysis of fluids physicochemical properties, particularly the liquids viscosity, is important for the optimization of industrial processes and products. For that, several empirical and semi-empirical equations have been proposed in the literature to study the viscosity of pure liquids and binary liquid mixtures. In this context, Messaâdi *et al.* have recently proposed an equation correlating the two parameters of the viscosity Arrhenius-type equation, such as the activation energy (E_a) and the pre-exponential factor (A_s), for 75 pure solvents. This paper aims to extend their model to binary liquid mixtures. To achieve this purpose, statistical analysis is made using data sets from the literature of some solvent binary mixtures at different compositions and temperatures.

The validation of the extended Messaâdi-Dhouibi equation for binary liquid mixtures simplifies the estimation of viscous behavior and the ensuing calculations. It is very useful for engineering data which permits to estimate one non-available parameter when the second one is available.

Keywords: Binary mixture; viscosity; Arrhenius behavior; Correlation; Statistics; Modeling.

Introduction

Viscosity is one of the most important physicochemical properties of fluids useful for the design and optimization of industrial processes such as chemical and food industry, cosmetics and pharmaceuticals, etc., viscosity is important for hydraulic calculations of fluid transport and for energy transference computation¹⁻⁶.

Nevertheless, the theoretical description of viscosity is quite complex. For that, several equations have been proposed in the literature essentially following Eyring theory^{1,2,7-11}. In addition, the excess thermodynamic functions (like the enthalpy of hydration) and the deviations of analogous non-thermodynamic functions (like the viscosity) of binary liquid mixtures are fundamental for understanding different types of intermolecular interactions in these mixtures.

Recently, several studies have tried to get more information about the internal structure of the binary liquid systems, particularly for binary mixtures composed of liquids having biological importance or promise^{1,12-30}. Thus, based on previous works, we have extended the study of viscosity Arrhenius behavior along with a new parameter labeled Arrhenius temperature (T_A) for some binary mixtures in different temperature ranges^{12-14,23-28}. In addition, knowing that the activation energy is a thermodynamic magnitude, partial molar activation energies have been calculated in order to release eventual contribution of individual interaction for each pure component within the mixture composition. Hence, the new introduced parameter defined as the current Arrhenius temperature (T_{Ai}) which is related to each component (i), permits to estimate the pure components boiling point constituting the assumed binary liquid mixture^{12,13,23-28}.

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Also, in some earlier papers^{14–20,27,28}, physicochemical excess properties are fitted to the reduced Redlich-Kister (R-K) equation and to the Herráez equation^{1,20–21}. Furthermore, we have found that the Herráez exponential form converges strictly to 0.5, 1, 1.5 or 2 which are independent of temperature^{1,2} and labeled Herráez universal exponent. In this context, we have compared these equations at different temperatures, and studied the effect of the use of different more or less parameters. Moreover, the reduced R-K function which is relative to the properties of thermodynamic physicochemical excess, equivalent to the apparent molar properties, turn out to be more sensitive than the purely excess properties to interactions, and hence tend to yield more information^{14–20, 27–28}.

Recently, an empirical equation has been proposed by Messaâdi *et al.*⁴² for modeling the relationship between the two parameters of viscosity Arrhenius-type equation for some pure classical solvents, as knowing the Arrhenius energy (E_a) and the factor ($\ln A_s$). The current work aims to extend the validity of the model to binary liquid mixtures. To that end, statistical methods are applied using data sets from the literature of liquid mixtures at different compositions and temperature ranges^{23–29}. This study is very useful for engineering data. Indeed, first, if the Messaâdi proposed equation is validated for binary liquid mixtures, the Arrhenius equation can be rewritten based on only one parameter instead of two ones and thus permits to simplify the engineering manipulations. Second, it will be possible to estimate with good accuracy one non-available parameter if the second one is available. The advantages and benefits of such scheme are obvious to the computational aspects in a myriad of disciplines in engineering and science.

Note also that this study will open new interesting field of profitable investigations such as the study of

$$\ln \eta = A + \frac{B}{T+c} + a \cdot \log T + b \cdot T + c \cdot T^2 + d \cdot T^p + \frac{D}{T^2} + \frac{E}{T^3} + \frac{F}{T^n} \quad (1)$$

where T is the absolute temperature and the rest of symbols are the models' parameters which at least, two are non-null for each previous equation and differ from one model to another.

Generally, we can classify the viscosity-temperature dependence into two main families for liquid systems according to their linear or non linear behavior when we plot the logarithm of shear viscosity ($\ln \eta$) against the reciprocal of absolute temperature ($1/T$).

Also, some additional multi-constant equations (Eq. (1)) are proposed for fluids deviating strongly to the Arrhenius behavior. We can cite for instance the melting salts, glasses and metals, ionic liquids, heavy and vegetable oils, fuels and biofuels, etc.^{30–31}. Otherwise, for the non linear behavior, it is found that the temperature dependence of dynamic

specific groups or families of organic liquids solvents, which will be very useful in large domains of applied chemistry and engineering. It could also pave the way to investigate more accurate values of the equation's parameters when the natures of fluids are classified differently.

Literature review

Due to the complex aspect of fluids, several theoretical methods for estimating viscosity are proposed in the literature. Among these theories, we can cite the distribution function theory proposed by Kirkwood *et al.*³², the molecular dynamic approach reported by Cumming and Evans³³ and the reaction rate theory of Eyring^{34–37}. In addition, the temperature dependence of the internal frictional coefficient of fluids is a complicated mathematical function; it is determined both by the thermal changes of the free volume and by thermodynamics of intermolecular interactions. So, the experimentalists express the temperature dependence of the dynamic viscosity by means of two-parameters frequently or more. Generally, empirical and semi-empirical methods provide reasonable results but they lack generality in approach, especially near or above the boiling temperature³⁰. Hence, experimental data available in literature show that the liquid viscosity decreases with temperature in non-linear and concave fashion, and it is slightly dependent on low pressure.

Numerous expressions have been proposed for representing the liquid viscosity (η) us a function of temperature (T) through available experimental data for an interpolation purpose^{38–59}. Varying between 2 and 5 constants by model, the most different forms of temperature-dependence of viscosity proposed previously in the context of the correlation' methods can be summarized in a global multi-constant equation as follow³¹:

viscosity can be fitted frequently with the Vogel-Fulcher-Tamman-type equation^{31,37–39} as following:

$$\ln \eta = \ln A_0 + \frac{E_0}{R} \left(\frac{1}{T - T_0} \right) \quad (2)$$

where A_0 , E_0 and T_0 are constants.

In addition, for the linear behavior, it is found that the temperature dependence of dynamic viscosity can be fitted frequently with the Arrhenius-type equation for numerous classic solvents, which can be rewritten in the logarithmic form:

$$\ln \eta = \ln A_s + \frac{E_a}{R} \left(\frac{1}{T} \right) \quad (3)$$

Where R , E_a and A_s are the gas constant, the Arrhenius activation energy and the pre-exponential (entropic) factor of the Arrhenius equation for the liquid system respectively.

Thus, the relationship between the logarithm of shear viscosity ($\ln \eta$) and the reciprocal of absolute temperature ($1/T$) for numerous liquid systems is assumed to be linear. In addition, the Arrhenius parameters, which are the activation energy (E_a) and the pre-exponential factor ($\ln A_s$), are thus independent of temperature.

Based on graphical projections, Messaâdi *et al.*³¹ determined these two main parameters i.e., the slope of the straight line is (E_a/R) and the intercept point is ($\ln A_s$). Also, they define and an additional third parameter (T_A) called the Arrhenius temperature deduced from the intercept with the abscissa axis (Eq. (4)).

$$T_A = \frac{-E_a}{R \ln(A_s)} \quad (4)$$

The viscosity-temperature dependence following the Eyring^{5,30,34–37} form can thus be rewritten as following:

$$\ln(\eta) = \frac{E_a}{R} \left(\frac{1}{T} - \frac{1}{T_A} \right) \quad (5)$$

In addition, the Fig. 1 showing nonlinear strong correlation between the Arrhenius activation energy E_a (kJ·mol⁻¹) and the Arrhenius temperature T_A (K), permit us to conclude that probably the Arrhenius temperature (T_A) and the activation energy (E_a) tend toward zero at the same time, and (T_A) cannot exceed a certain limit value (T_{Alim}) when takes infinite value. Indeed, it's clearly shown by the general trend and shape of (Fig. 1) and the mathematical expression of Eq. 6 for which ($T_{Alim} = T_0 = 288.65 \pm 4.38$) K³¹.

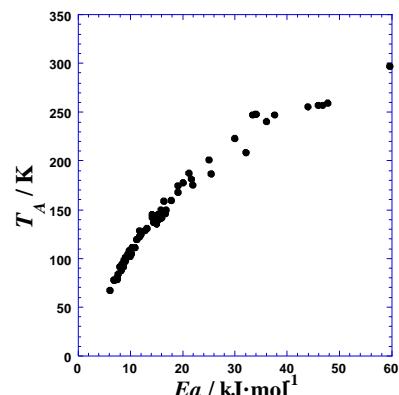


Figure 1. Scatter plots for pair-wise correlations between Arrhenius parameters.

$$T_A = [1 - e^{-\frac{E_a}{\alpha_0 R T_0}}] \cdot T_0 \quad (6)$$

Where $R = 8.3144721$ J·mol⁻¹, (T_0) K and (α_0) are empirical adjustable parameters.

Based on statistical and econometric analysis^{50,60}, Messaâdi *et al.*³¹ found significant statistical correlation between the two parameters of viscosity Arrhenius-type equation as knowing the entropic factor ($\ln A_s$)_{exp} and the activation energy (E_a)_{exp}.

Thus, they proposed the Eq. (8) and Eq. (9) to model this relationship.

$$E_a = -\alpha_0 R T_0 \cdot \ln(1 - \frac{T_A}{T_0}) \quad (7)$$

$$\ln(A_s) = \frac{-E_a}{R T_0 \cdot [1 - e^{-\frac{E_a}{\alpha_0 R T_0}}]} \quad (8)$$

$$E_a = -\frac{R T_0 \cdot [\ln(A_s) + \alpha_0]}{[1 - e^{-\frac{R T_0 \cdot \ln(A_s)}{\gamma_0}}]} \quad (9)$$

where ($T_0 = 288.65 \pm 4.38$) K, ($\alpha_0 = 9.0208 \pm 0.5370$) and ($\gamma_0 = (37.49 \pm 5.62) \times 10^3$).

Note that the parameters (T_0 , α_0 and γ_0) are calculated based on the overall mean's boiling temperatures of the binary solvents mixtures presented in Table 3. Nevertheless, their values may differ slightly if they are calculated for specific families of solvents separately such as organic versus inorganic solvents, polar versus non-polar liquids, high versus low viscosity liquids, etc., which may enhance the precision during their engineering use.

Replacing the parameters' optimal values of (T_0 , α_0 and γ_0) in Eq. (8) and Eq. (9), the two following equations become practical to deduce one of the two Arrhenius parameters if it is absent or unavailable using the second one.

Where the parameters (T_0 , α_0 and γ_0) take into consideration the global average of the boiling temperatures of the studied set of solvents and can slightly deviate from the given values when the statistical investigation is restricted to specific families of solvents such as organic versus inorganic solvents, polar versus non-polar liquids, high versus low viscosity liquids, etc.

In instance, the two following practical equations can be used to deduce one of the two

Arrhenius parameters if it is absent or non-available using the second one.

$$\ln(A_s) = \frac{-E_a}{2400 \cdot [1 - e^{-\frac{E_a}{21650}}]} \quad (10)$$

$$E_a = -\frac{2400 \cdot [\ln(A_s) + 9.0208]}{[1 - e^{0.064 \cdot \ln(A_s)}]} \quad (11)$$

Consequently, the Messaâdi *et al.* expressions³¹ allow redefining the Arrhenius equation (Eq. (3)) by using a single parameter (Eq. (12) or Eq. (13)) instead of two ones (Eq. (3)) are the following:

Consequently, the Messaâdi–Dhouibi expressions⁴² allow redefining the Arrhenius equation by using a single parameter (Eq. (12) or Eq. (13)) instead of two ones (Eq. (3)) are the following:

$$\ln(\eta) = \ln(A_s) - \frac{RT_0[\ln(A_s) + \alpha_0]}{R\gamma_0} \cdot \left(\frac{1}{T} \right) \quad (12)$$

$$\ln(\eta) = \frac{-E_a}{RT_0[1-e^{-\alpha_0 RT_0}]} + \frac{E_a}{R} \cdot \left(\frac{1}{T} \right) \quad (13)$$

Finally, for numerical and practical use of Eq. (12) or Eq. (13), Messaâdi *et al.*³¹ proposed the following simplified practical equations with good approximation for numerous classical solvents obeying to the viscosity Arrhenius-type equation (Eq. (3)) as follows:

$$\ln(\eta) = \ln(A_s) - \frac{2400[\ln(A_s) + 9.0208]}{[1-e^{0.064 \cdot \ln(A_s)}]} \cdot \left(\frac{1}{T} \right) \quad (14)$$

$$\ln(\eta) = \frac{-E_a}{2400[1-e^{-\frac{E_a}{2400}}]} + \frac{E_a}{8.31447} \cdot \left(\frac{1}{T} \right) \quad (15)$$

Empirical validation for binary mixtures

Assuming the validity of the Messaâdi-Dhouibi equations, Eq. (8) and Eq. (9), modeling the relationship between the two parameters of viscosity Arrhenius-type equation for pure liquid, this paper aims to study and validate their eventual extension

from pure to binary fluid mixtures. For that, we use 241 experimental data on viscosity for 13 binary liquid mixtures, over different temperature ranges at atmospheric pressure. From literature experimental studies of viscosity-temperature dependence in some binary fluid mixtures, and through linear regression using (Eq. (3)), Table 3 in Appendix section presents the calculated values of the entropic factor ($\ln As$)_{exp}, the activation energy (Ea)_{exp}, the Arrhenius activation temperature ($T^* = Ea / R$) and the Arrhenius temperature (T_A in (Eq. (4)), as defined by Messaâdi *et al.*³¹.

Our study will be based on the analysis of the quality of approximation of the entropic factor ($\ln As$) and the activation energy (Ea) using the Messaâdi-Dhouibi equations and the new data set. Thus, we have estimated the entropic factor, ($\ln As$)_{est}, by replacing the experimental activation energy (Ea)_{exp} of binary liquid mixtures data in Eq. (9). In addition, we have estimated the experimental activation energy (Ea)_{est} by replacing the experimental entropic factor($\ln As$)_{exp} in Eq. (8). Table 1 presents descriptive statistics on experimental and estimated values for both parameters.

Table 1. Descriptive statistics on experimental and estimated values of E_a and $\ln As$.

Parameters	Mean	σ	Min	Max
(E_a) _{exp}	17.236	8.084	9.053	46.763
(E_a) _{est}	17.088	6.925	8.470	40.903
($\ln As$) _{exp}	-13.217	2.209	-21.857	-10.780
($\ln As$) _{est}	-13.178	2.261	-22.025	-11.038

The descriptive statistics show clearly that the experimental data are almost similar to the corresponding Ea -values and $\ln As$ -values estimated from Eq. (8) and Eq. (9) regardless of the calculated statistical indicators. This permits to expect a good estimation accuracy of the Messaâdi equations for binary liquids mixture. Nevertheless, to confirm this

result, there is a need to apply adequate statistical tests of comparison of populations. For that, we use the Wilcoxon Signed-Rank test⁶⁰ where the null hypothesis assumes that the two samples are from populations with the same distribution and thus we can decide whether the corresponding data population distributions are identical (Table 2).

Table 2. Result of the Wilcoxon Signed-Rank test for both parameters.

Variables	z	Prob> z
(E_a) _{exp} versus (E_a) _{est}	-0.905	0.365
($\ln As$) _{exp} versus ($\ln As$) _{est}	-1.643	0.101

Results of the Wilcoxon Signed-Rank test lead to accept the null hypothesis for both parameters E_a and $\ln As$ which confirms that statistically the experimental and the estimated distributions are significantly the same and proves the predictive power of Messaâdi equations for binary liquid mixtures in addition to pure liquids.

This result can be shown graphically based on Fig. 2 and Fig. 3 which compare the experimental versus the estimated values for both parameters. Indeed, Fig. 2 shows the experimental data of one parameter on x -axis with experimental and estimated values simultaneously of the second parameter on the y -axis. However, Fig. 3 shows the experimental

values of the activation energy (E_a)_{exp} and the entropic factor ($\ln As$)_{exp} in direct comparison with the estimated values (E_a)_{exp} and ($\ln As$)_{calc}, respectively. Thus, based on these figures, we can deduce that the gap between experimental and estimated values is indicating a slight discrepancy and confirms the good quality of approximation of the Messaâdi equations for binary liquid mixtures.

In addition, Fig. 4 shows the normalized estimation errors of both parameters. The figures confirm also the precedent result about the good quality of estimators. Indeed, it appears clearly that the normalized estimation errors are in close to zero for both parameters.

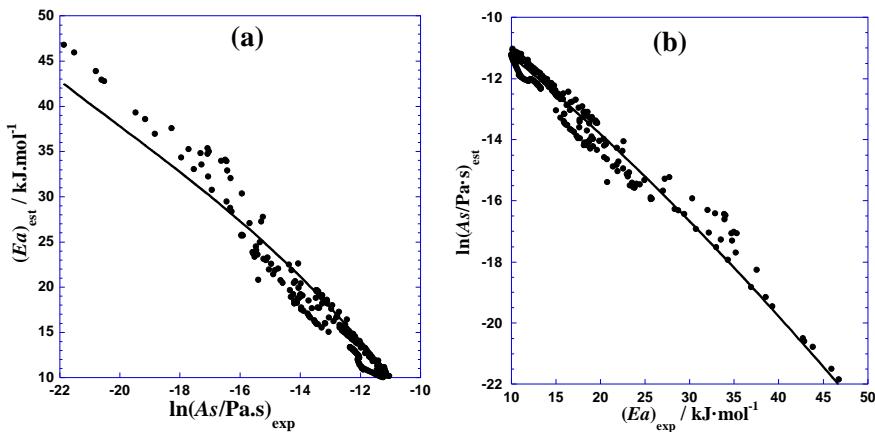


Figure 2. (a) . Comparison between the experimental entropic factor $(\ln As)_{exp}$ and the estimated ones $(\ln As)_{est}$ as function of the experimental entropic factor $(Ea)_{exp}$; (b): Comparison between the experimental activation energy $(Ea)_{exp}$ and the estimated ones $(Ea)_{est}$ as function of the experimental entropic factor $(\ln As)_{exp}$. (●): $(\ln As)_{exp}$; (—): $(\ln As)_{est}$.

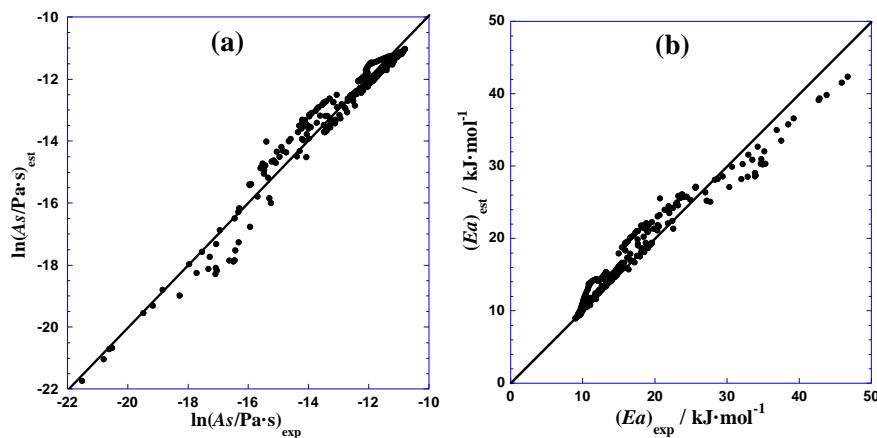


Figure 3. (a) . Comparison between the experimental entropic factor $(\ln As)_{exp}$ and the estimated ones $(\ln As)_{est}$ from Eq. (8) ; (b): Comparison between the experimental activation energy $(Ea)_{exp}$ and the estimated ones $(Ea)_{est}$ from Eq. (9).

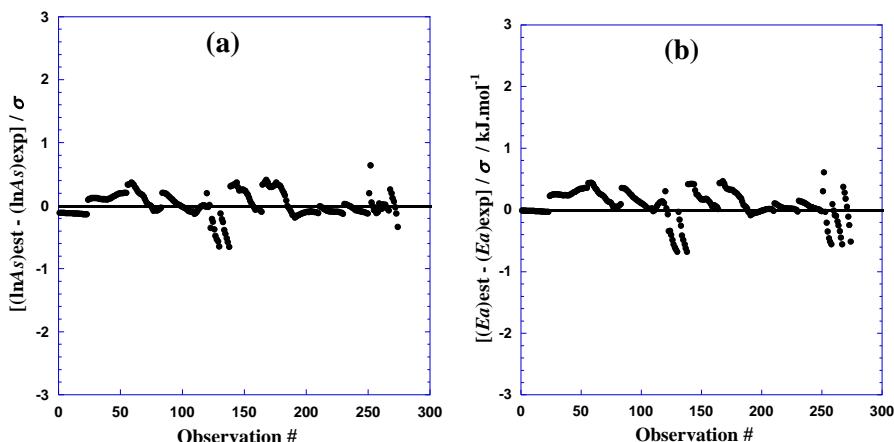


Figure 4. Normalized deviation plot related to: (a): the entropic factor: $[(\ln As)_{exp} - (\ln As)_{est}] / \sigma$ and (b): the activation energy: $[(Ea)_{exp} - (Ea)_{est}] / \sigma$ against the number of observations (binary mixture).

Conclusion

Using the Messaâdi-Dhouibi equations, Eq. (7) and Eq. (8), which model the relationship between the two parameters of viscosity Arrhenius-type equation for some Newtonian pure liquids⁴², as knowing the entropic factor ($\ln A_s$) and the activation energy (E_a), the goal of the present work, was to test the quality of their approximation power for some Newtonian binary liquid mixtures. For that, a sample of 241 experimental data on viscosities for 13 binary liquid mixtures over different temperature ranges at atmospheric pressure is used to ($\ln A_s$) and (E_a) values using the Messaâdi-Dhouibi equations. Then, based on the result of a statistical test, as knowing the Wilcoxon Signed-Rank test, we found that the experimental and the estimated distributions are statistically and significantly the same which prove the good predictive power of Messaâdi-Dhouibi equations for binary liquid mixtures in addition to pure liquids.

This result is important in fluids engineering since the validation of the extended Messaâdi equation for binary liquid mixtures simplifies the estimation of viscous behavior and the ensuing calculations by reducing the number of viscosity equation parameters to become depending on only one parameter instead of two, which permit to simplify the engineering manipulations. In addition, it will be possible to estimate with good accuracy one non-available parameter if the second one is available. The advantages and benefits of such scheme are obvious to the computational aspects in a myriad of disciplines in engineering and science.

Note that the proposed equation presents good concordance also for low and moderate viscous fluids which have not got very high values of activation energy ($5 < E_a < 60 \text{ kJ}\cdot\text{mol}^{-1}$) and not very low values of pre-experimental factor ($-25 < \ln(A_s / \text{Pa}\cdot\text{s}) < -9$).

We hope that this study will open new interesting field of profitable investigations such as the study of specific groups or families of organic liquids solvents (alcohols, ketones, amides etc.). It could also pave the way to estimate more accurate values of the proposed equation's parameters, when the natures of fluids are classified separately (high, moderate, low viscous liquids) or solvent characteristics (polar, non-polar etc.). Hence, each separated investigation can provide specific parameter values and it leads to more accurate specific ones with better statistical results, demonstrating how this model deserves validity. In the same way, this correlation can encourage theorists to combine or merge some previous distinct already available theories. In addition, we hope that it will be useful in large domains of applied chemistry and engineering.

Finally, note that an additional study on the eventual relationship between the Arrhenius parameters and the properties of great number liquid

systems can prove how the method predicts the properties of other non-treated fluid systems. In order to tightly find the utility of the Arrhenius temperature and develop a means for estimating such quantities, more binary liquid mixtures will be studied against composition in future to give a more clear discussed protocol.

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Appendix

Table 3 Experimental data on entropic factor ($\ln As_{\exp}$), activation energy (Ea_{\exp}), Arrhenius activation temperature (T^*/K) and the Arrhenius temperature (T_A/K) of 241 observations for 13 binary liquid mixtures (i) over different temperature ranges $x(i)$ at atmospheric pressure at different molar fraction on x .

1,4-Butanediol (x) + water ($1-x$) ³⁶					0.6404	1999.77	-12.7924	16.627	156.33
x (1)	T^*	$\ln As$	Ea	T_A	0.7953	1744.43	-12.1618	14.504	143.44
0	1918.34	-13.1615	15.950	145.75	0.9529	1427.39	-11.3956	11.868	125.26
0.100	2629.25	-14.2775	21.861	184.15	1	1337.43	-11.1996	11.120	119.42
0.200	3458.34	-16.3164	28.754	211.95	Methanol (x) + DMF ($1-x$) ³⁰				
0.300	3337.23	-15.2366	27.747	219.03	x (9)	T^*	$\ln As$	Ea	T_A
0.400	4033.40	-17.2713	33.536	233.53	0	1325.20	-11.9354	11.018	111.03
0.500	4235.63	-17.7063	35.217	239.22	0.0101	1325.20	-11.9307	11.018	111.07
0.600	4184.50	-17.3094	34.792	241.75	0.0202	1325.10	-11.9251	11.018	111.12
0.700	4174.39	-17.0689	34.708	244.56	0.0505	1323.10	-11.9035	11.001	111.15
0.800	4205.86	-17.0310	34.969	246.95	0.0995	1316.20	-11.8568	10.944	111.01
0.900	4249.79	-17.0732	35.335	248.92	0.1515	1303.20	-11.7884	10.835	110.55
1	4093.27	-16.4848	34.033	248.31	0.2004	1288.20	-11.7153	10.711	109.96
1,2-Butanediol (x) + pentanol ($1-x$) ³⁶					0.2525	1274.10	-11.6438	10.594	109.42
x (2)	T^*	$\ln As$	Ea	T_A	0.3004	1261.60	-11.5798	10.490	108.95
0	2452.17	-13.9834	20.388	175.36	0.3535	1246.41	-11.5050	10.363	108.34
0.200	2703.94	-14.3732	22.482	188.12	0.3993	1236.30	-11.4513	10.279	107.96
0.400	3254.07	-15.6810	27.056	207.52	0.4545	1225.80	-11.3926	10.192	107.60
0.500	3695.41	-16.9298	30.725	218.28	0.5008	1216.81	-11.3435	10.117	107.27
0.600	3970.76	-17.5264	33.015	226.56	0.5556	1213.01	-11.3093	10.086	107.26
0.700	4442.75	-18.8294	36.939	235.95	0.5728	1212.90	-11.3023	10.085	107.31
0.800	4726.13	-19.4663	39.295	242.79	0.6566	1205.30	-11.2470	10.021	107.17
0.900	5157.44	-20.5991	42.881	250.37	0.6998	1203.60	-11.2265	10.007	107.21
1	5624.27	-21.8569	46.763	257.32	0.7576	1197.60	-11.1877	9.957	107.05
1,2-Butanediol (x) + water ($1-x$) ³⁶					0.8014	1190.80	-11.1513	9.901	106.79
x (3)	T^*	$\ln As$	Ea	T_A	0.8586	1183.60	-11.1093	9.841	106.54
0	1920.75	-13.1672	15.970	145.87	0.8996	1175.50	-11.0693	9.774	106.19
0.100	2497.72	-15.3923	20.767	162.27	0.9596	1153.70	-10.9784	9.592	105.09
0.200	3410.37	-16.2775	28.355	209.51	0.9798	1146.10	-10.9469	9.529	104.70
0.400	4125.99	-17.9441	34.305	229.94	0.9899	1142.80	-10.9326	9.502	104.53
0.600	4636.52	-19.1528	38.550	242.08	1	1140.20	-10.9208	9.480	104.41
0.800	5140.61	-20.5073	42.741	250.67	DMA (x) + formamide ($1-x$) [31]				
0.900	5275.31	-20.7865	43.861	253.79	x (10)	T^*	$\ln As$	Ea	T_A
1	5524.51	-21.5095	45.933	256.84	0	1973.67	-12.4418	16.410	158.63
1,4-Butanediol (x) + pentanol ($1-x$) ³⁶					0.0350	2074.70	-12.7004	17.250	163.36
x (4)	T^*	$\ln As$	Ea	T_A	0.0700	2161.29	-12.9235	17.970	167.24
0	2209.07	-13.0977	18.367	168.66	0.1110	2233.46	-13.1026	18.570	170.46
0.200	2716.75	-14.0569	22.588	193.27	0.1500	2293.59	-13.2616	19.070	172.95
0.400	3274.70	-15.2840	27.227	214.26	0.2053	2341.70	-13.3902	19.470	174.88
0.600	3649.05	-15.9274	30.340	229.10	0.2414	2359.74	-13.4440	19.620	175.52
0.700	3852.55	-16.3093	32.032	236.22	0.3009	2363.35	-13.4649	19.650	175.52
0.800	3955.00	-16.4205	32.884	240.86	0.3563	2347.71	-13.4509	19.520	174.54
0.900	4082.05	-16.6168	33.940	245.66	0.4177	2296.00	-13.3455	19.090	172.04
1	4077.69	-16.4379	33.904	248.07	0.4580	2252.70	-13.2616	18.730	169.87
Propylene Glycol (x) + water ($1-x$) ³⁵					0.5011	2182.94	-13.0977	18.150	166.67
x (5)	T^*	$\ln As$	Ea	T_A	0.5474	2113.18	-12.9611	17.570	163.04
0	1808.10	-13.0499	15.033	138.55	0.5972	2021.78	-12.7612	16.810	158.43
0.300	2392.69	-14.0471	19.894	170.33	0.6500	1906.32	-12.4991	15.850	152.52
0.500	3000.07	-15.3295	24.944	195.71	0.7092	1781.23	-12.2324	14.810	145.62
0.700	3540.45	-16.4383	29.437	215.38	0.7500	1687.42	-12.0238	14.030	140.34
1	4516.33	-18.2660	37.551	247.25	0.7948	1594.81	-11.8290	13.260	134.82
DMA (x) + DMF ($1-x$) ²⁷					0.8910	1407.19	-11.4341	11.700	123.07

x (6)	T^*	lnAs	Ea	T_A	1	1188.77	-10.9648	9.884	108.42
0	1088.82	-10.7798	9.053	101.01		DMA (x) + 2-ethoxyethanol (1- x) ³²			
0.0200	1090.87	-10.7836	9.070	101.16	0.000	1900.66	-12.6820	15.803	149.87
0.0500	1094.12	-10.7895	9.097	101.41	0.0500	1879.01	-12.6211	15.623	148.88
0.1062	1100.13	-10.8003	9.147	101.86	0.1042	1839.56	-12.5030	15.295	147.13
0.1500	1104.46	-10.8079	9.183	102.19	0.1500	1801.32	-12.3894	14.977	145.39
0.2171	1110.95	-10.8187	9.237	102.69	0.2075	1756.94	-12.2623	14.608	143.28
0.2600	1115.16	-10.8262	9.272	103.01	0.2500	1726.39	-12.1783	14.354	141.76
0.3094	1119.61	-10.8330	9.309	103.35	0.3097	1686.46	-12.0732	14.022	139.69
0.350	1123.22	-10.8386	9.339	103.63	0.3650	1651.34	-11.9852	13.730	137.78
0.4050	1128.03	-10.8464	9.379	104.00	0.4111	1623.31	-11.9184	13.497	136.20
0.4550	1132.12	-10.8522	9.413	104.32	0.4600	1594.81	-11.8535	13.260	134.54
0.5042	1135.97	-10.8572	9.445	104.63	0.5115	1565.58	-11.7899	13.017	132.79
0.5500	1139.58	-10.8623	9.475	104.91	0.5650	1534.07	-11.7230	12.755	130.86
0.6071	1144.03	-10.8685	9.512	105.26	0.6110	1506.05	-11.6646	12.522	129.11
0.6550	1147.76	-10.8738	9.543	105.55	0.6550	1477.79	-11.6051	12.287	127.34
0.7140	1152.09	-10.8790	9.579	105.90	0.7096	1440.74	-11.5266	11.979	124.99
0.7500	1154.61	-10.8824	9.600	106.10	0.7550	1407.79	-11.4556	11.705	122.89
0.7968	1157.50	-10.8849	9.624	106.34	0.8073	1365.33	-11.3604	11.352	120.18
0.8500	1160.99	-10.8886	9.653	106.62	0.8500	1329.25	-11.2794	11.052	117.85
0.9112	1164.84	-10.8925	9.685	106.94	0.9041	1280.30	-11.1691	10.645	114.63
0.9500	1167.12	-10.8943	9.704	107.13	1	1173.05	-10.9119	9.753	107.50
0.9800	1168.69	-10.8952	9.717	107.27		DMA (x) + water (1- x) ³³			
1	1169.77	-10.8957	9.726	107.36		Methanol (x) + DMA (1- x) ²⁸			
x (7)	T^*	lnAs	Ea	T_A	x (12)	T^*	lnAs	Ea	T_A
0	1556.44	-12.1720	12.941	127.87	0.000	1865.39	-13.2841	15.510	140.42
0.0202	1574.60	-12.2330	13.092	128.72	0.0020	1910.91	-13.4027	15.888	142.58
0.0404	1588.44	-12.2780	13.207	129.37	0.0100	1961.20	-13.5267	16.306	144.99
0.0707	1600.46	-12.3190	13.307	129.92	0.0150	2051.51	-13.7537	17.057	149.16
0.1010	1604.31	-12.3340	13.339	130.07	0.0250	2131.83	-13.9575	17.725	152.74
0.1250	1604.31	-12.3370	13.339	130.04	0.0650	2271.28	-14.2976	18.885	158.86
0.1414	1599.74	-12.3250	13.301	129.80	0.1011	2493.29	-14.6434	20.730	170.27
0.1717	1591.32	-12.3020	13.231	129.35	0.1200	2626.41	-14.8364	21.837	177.02
0.2020	1580.02	-12.2710	13.137	128.76	0.1495	2778.40	-15.2132	23.101	182.63
0.2320	1567.15	-12.2370	13.030	128.07	0.2004	2891.85	-15.4562	24.044	187.10
0.2727	1548.14	-12.1850	12.872	127.05	0.2512	3088.28	-15.9504	25.677	193.62
0.3030	1534.07	-12.1490	12.755	126.27	0.3002	3099.49	-15.9349	25.771	194.51
0.3240	1521.80	-12.1150	12.653	125.61	0.3501	3087.22	-15.8946	25.669	194.23
0.3535	1508.70	-12.0960	12.544	124.73	0.4009	2941.29	-15.4684	24.455	190.15
0.3838	1500.76	-12.0710	12.478	124.33	0.5018	2795.33	-15.0758	23.242	185.42
0.4050	1491.62	-12.0550	12.402	123.73	0.6031	2458.46	-14.2065	20.441	173.05
0.4444	1481.63	-12.0370	12.319	123.09	0.6505	2128.02	-13.3576	17.693	159.31
0.4950	1470.09	-12.0260	12.223	122.24	0.7005	1950.76	-12.8787	16.220	151.47
0.5455	1462.03	-12.0290	12.156	121.54	0.7550	1818.54	-12.5424	15.120	144.99
0.5960	1456.26	-12.0410	12.108	120.94	0.7550	1664.58	-12.1411	13.840	137.10
0.6440	1452.17	-12.0640	12.074	120.37	0.8048	1519.92	-11.7566	12.637	129.28
0.6869	1446.39	-12.0700	12.026	119.83	0.8501	1477.09	-11.6942	12.281	126.31
0.7273	1440.02	-12.0790	11.973	119.22	0.9049	1419.42	-11.5909	11.802	122.46
0.7778	1428.35	-12.0800	11.876	118.24	0.9400	1357.21	-11.4368	11.285	118.67
0.8283	1411.15	-12.0650	11.733	116.96	0.9700	1329.90	-11.2725	10.758	114.78
0.8788	1387.58	-12.0340	11.537	115.31	1	1222.64	-11.0782	10.166	110.36
x (13)	T^*	lnAs	Ea	T_A		1,4-dioxane (x) + water (1- x) ²⁹			
0.9150	1369.18	-12.0170	11.384	113.94		1925.56	-13.4756	16.010	142.89
0.9390	1351.86	-11.9800	11.240	112.84		1944.80	-13.5125	16.170	143.93
0.9610	1340.67	-11.9540	11.147	112.15		1997.72	-13.6551	16.610	146.30
0.9798	1324.68	-11.9340	11.014	111.00		2084.32	-13.8827	17.330	150.14
0.9899	1317.82	-11.9230	10.957	110.53		2191.36	-14.1019	18.220	155.40
x (8)	T^*	lnAs	Ea	T_A	0.0530	2229.85	-14.1193	18.540	157.93
					0.0737	2249.09	-14.0782	18.700	159.76

0	1914.77	-13.4448	15.920	142.41	0.0841	2255.10	-14.0538	18.750	160.46
0.0021	2017.93	-13.6988	16.778	147.31	0.1114	2270.74	-14.0055	18.880	162.13
0.005	2104.76	-13.9578	17.500	150.80	0.1433	2286.38	-13.9863	19.010	163.47
0.0106	2183.18	-14.1946	18.152	153.80	0.1726	2300.81	-13.9875	19.130	164.49
0.0222	2219.74	-14.1921	18.456	156.41	0.2065	2306.82	-13.9792	19.180	165.02
0.0348	2226.60	-14.0895	18.513	158.03	0.2501	2288.78	-13.9098	19.030	164.54
0.0486	2306.70	-14.2196	19.179	162.22	0.3029	2226.24	-13.7157	18.510	162.31
0.0545	2360.46	-14.3414	19.626	164.59	0.3560	2128.82	-13.4201	17.700	158.63
0.0638	2457.88	-14.5792	20.436	168.59	0.4203	1998.92	-13.0385	16.620	153.31
0.0737	2575.27	-14.8845	21.412	173.02	0.4996	1865.42	-12.6835	15.510	147.08
0.0806	2638.53	-15.0404	21.938	175.43	0.5367	1825.73	-12.5719	15.180	145.22
0.0915	2804.51	-15.5085	23.318	180.84	0.5999	1760.79	-12.4116	14.640	141.87
0.1114	2851.29	-15.5609	23.707	183.24	0.6479	1707.87	-12.2750	14.200	139.13
0.1200	2867.41	-15.5823	23.841	184.02	0.7309	1599.62	-11.9742	13.300	133.59
0.1290	2868.49	-15.5590	23.850	184.36	0.7621	1559.93	-11.8019	12.970	132.18
0.1433	2834.22	-15.4155	23.565	183.86	0.7953	1523.85	-11.7641	12.670	129.53
0.1698	2761.57	-15.1394	22.961	182.41	0.8686	1472.13	-11.6349	12.240	126.53
0.1999	2712.50	-14.9522	22.553	181.41	0.9092	1454.09	-11.5958	12.090	125.40
0.2347	2650.08	-14.7246	22.034	179.98	0.9760	1461.31	-11.6436	12.150	125.50
0.3230	2480.37	-14.1545	20.623	175.24	0.9855	1476.94	-11.7024	12.280	126.21
0.4499	2257.99	-13.4707	18.774	167.62	1	1521.44	-11.8344	12.650	128.56
0.4996	2182.34	-13.2567	18.145	164.62					

*N,N-dimethylacetamide: (DMA) , N,N-dimethylformamide: (DMF).