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Analysis of correlation between viscosity Arrhenius parameters: Extension to ternary liquid mixtures

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Abstract: The study of viscosity-temperature dependence is important for the design and the optimization of several industrial processes and products. In this context, an interesting equation is recently proposed for pure solvents correlating the two parameters of the viscosity Arrhenius-type equation, as knowing the activation energy (*Ea*) and the pre-exponential factor (*As*), allowing the simplification of the viscosity Arrhenius equation to become an expression depending on only one parameter instead of two. The present work investigates the validity of the simplified Arrhenius-type equation for ternary fluids mixtures. The extension of the proposed equation to ternary liquid mixtures is very important since it simplifies the estimation of viscous behavior and the ensuing calculations. Using statistical methods and 114 experimental data from the literature on viscosity for 5 ternary liquid mixtures over different temperature ranges at atmospheric pressure, we found that the validation of the proposed equation depends significantly on the density of liquids and is validated only for 4 studied ternary liquid mixtures. This result opens the way to investigate the sensitivity of the equation's parameters for more specific mixtures.

Keywords: Viscosity; ternary mixture; Arrhenius temperature; correlation; statistics

Introduction

Fluids viscosity is one of the main transport property involved in chemical engineering and several industries such as food industry, cosmetics and pharmaceuticals, etc. Indeed, viscosity is essential for hydraulic calculations of fluid transport and for energy transference computation [1-6]. Consequently, several theoretical, semi-theoretical and empirical equations of fluid viscosity have been proposed in the literature following three main theories: the reaction rate theory of Eyring [7–9], the molecular dynamic approach proposed by Cumming and Evans [10] and the distribution function theory of Kirkwood *et al.* [11].

Particularly, several studies have been made in order to analyze the viscosity-temperature dependence. Thus, different expressions have been suggested in the literature for modeling the liquid viscosity (η) against temperature (T) through available experimental data for an interpolation aim [12-24].

In this context, Kacem et al. [25] have recently found a significant correlation between the two parameters of viscosity Arrhenius-type equation for some common solvents, which are the Arrhenius energy (*Ea*) and the factor (*ln*As). Hence, they have proposed an empirical equation modeling this relationship and then they have proposed a simplified expression of the Arrhenius-type equation.

The current work aims to extend the validity of the Kacem et al. [25] models to ternary mixtures. For that, statistical tools are applied using data sets from the literature of ternary liquid mixtures at different compositions and temperature ranges [26, 27].

The validation of the proposed equation is important since it allows redefining the Arrhenius equation by using single parameter instead of two. The advantages and benefits of such scheme are obvious to the computational aspects in a myriad of disciplines in engineering and science.

The temperature dependence of liquid viscosity: a literature review

Several equations have been proposed in the literature for representing the liquid viscosity (η) upon temperature (T) through experimental data for interpolation purpose. Generally, they differ according to the number of parameters. For that, one can classify them into three categories: two-constant, three-constant and multi-constants equation.

The representation of Newtonian liquid viscosity-temperature dependence with two parameters is proposed initially by Guzman [12] and known as the

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Andrade equation as following:

$$\eta = Ae^{\frac{B}{T}} \qquad (1)$$

where A and B are positive constants and are characteristics of each pure liquid.
Following the Andrade equation, many expressions have been suggested based on two parameters [13-15]. However, the most popular is the Arrhenius type-equation which may be expressed as following:

$$ln\,\eta = ln\,A_s + \frac{E_a}{R}(\frac{1}{T}) \qquad (2)$$

Where R, E_a and A_s are the perfect gas constant, the Arrhenius activation energy and the pre-exponential factor of the Arrhenius equation.

Also, considering the Newtonian liquids not obeying to the Arrhenius behavior, several non-linear models with two parameters are proposed which are ranging from semi-theoretical to purely empirical and discussed in previous works [14-24].

The three-constant representation of the viscosity-temperature dependence is proposed originally by Vogel [17] and known as Vogel-Fulcher-Tammann-type equation [17, 19]:

$$ln\,\eta = A - \frac{B}{T-C} \qquad (4)$$

Where A, B and C are constants.
Following Vogel, several researchers have proposed similar expressions of the viscosity-temperature dependence [14,15,20]. In addition, others researcher have proposed different forms using three parameters such as the Eq. 5 and Eq. 6, which are proposed by Girifalco [20] and Thorpe and Rodger [21] respectively.

$$\eta = \frac{C}{1+AT+BT^2} \qquad (5)$$

$$ln\eta = A + \frac{B}{T} + \frac{D}{T^2} \qquad (6)$$

where A, B and C are parameters' models.

Concerning the expressions based on more than three constants, they have been proposed in the aim to improve the accuracy of the representation, particularly over wider ranges of temperature. The most popular multi-constant equations are polynomials, such as the Eq. 7 [16] with 4 parameters and the Eq. 8 [24] with 5 parameters.

$$ln\eta = A + \frac{B}{T} + C.T + DT^2 \qquad (7)$$

$$ln\eta = A + \frac{B}{T} + C.lnT + DT^E \qquad (8)$$

where A, B, C D and E are the equation' parameters.

Recently, Kacem et al. [25] have shown a significant correlation between the two parameters of viscosity Arrhenius-type equation, the Arrhenius activation energy (Ea) and the pre-exponential factor (lnAs) for some pure solvents studied at different temperature ranges. Thus, they have proposed the following equations linking the two parameters:

$$E_a = R\times(-lnA_s)^{2.933} \qquad (9)$$

$$lnA_s = -(\frac{E_a}{R})^{0.341} \qquad (10)$$

where R is the gas constant.

Hence, according to Kacem et al. [25], the Arrhenius-type equation can be simplified to become an expression based on only one parameter instead of two ones as following:

$$ln\eta = lnA_s + \lambda\,(-lnA_s)^{2.933}\,(\frac{1}{T}) \qquad (11)$$

$$ln\,\eta = -(\frac{E_a}{R})^{0.341} + \frac{E_a}{R}.(\frac{1}{T}) \qquad (12)$$

Note that the Kacem et al. [25] expressions have been also validated for binary mixtures and some specific families of liquids such as alcohols, organic acids, aldehydes, ketones, alkanes, etc [28-31].

Empirical investigation: extension to ternary mixtures

Assuming the validation of the Kacem-Ouerfelli equations, Eq. (9) to Eq. (12), for pure and binary liquids mixture, our investigation will focus on analyzing its eventual extended validity to ternary fluid mixtures. For that, we use 114 experimental data from the literature on viscosity for 5 ternary liquid mixtures over different temperature ranges at atmospheric pressure [26, 27]. Thus, the proposed investigation will be based on analyzing the predictive power of Kacem-Ouerfelli equations by comparing the experimental with the estimated values of the Arrhenius-type equation parameters, as knowing the Arrhenius activation energy Ea (kJ·mol^{-1}) and the logarithm of the entropic factor of Arrhenius ln(As/Pa·s).

Table 1 presents the experimental values of Arrhenius activation energy Ea (kJ·mol^{-1}) and the logarithm of the entropic factor of Arrhenius ln(As/Pa·s). In addition, for the discussion, we report in the same Table other temperature parameters as defined by Kacem *et al.* [25], as knowing the Arrhenius temperature (T_A /K) and the Arrhenius activation temperature (T*/K, T* = Ea/R) [25, 28-30].

Table 1. Experimental values of Arrhenius activation energy Ea (kJ·mol-1), the logarithm of the entropic factor of Arrhenius ln(As/Pa·s), the Arrhenius temperature TA (K) and the Arrhenius activation temperature (T*= Ea/R /K) for 5 ternary liquid mixtures.

System	T*/K	ln*As*	*Ea*	*TA*/K
Mixture (1) [26] **Water (1) + ethane-1,2-diol (2) + methanol (3)**	1824	-12.3342	15.1656	147.8811
	1806.422	-12.2191	15.01944	147.8364
	1939.484	-12.5787	16.12579	154.1879
	1904.363	-12.4472	15.83377	152.9951
	1968.904	-12.7341	16.3704	154.6172
	1957.391	-12.7695	16.27468	153.2869
	1861.824	-12.5763	15.48009	148.0427
	1782.675	-12.4339	14.822	143.3727
	1593.071	-11.9574	13.24555	133.229
	1407.004	-11.534	11.69849	121.9874
	1904.53	-13.4136	15.83516	141.9849
Mixture (2) [26] **Water (1) + ethane-1,2-diol (2) + ethanol (3)**	2447.966	-13.8519	20.35354	176.7241
	2179.243	-13.0848	18.11926	166.5482
	2199.871	-13.1935	18.29077	166.739
	2167.775	-13.1521	18.0239	164.823
	2146.622	-13.2296	17.84803	162.2587
	2084.679	-13.157	17.33301	158.4463
	2060.626	-13.2316	17.13302	155.7355
	2045.117	-13.297	17.00407	153.803
	1928.62	-13.0536	16.03545	147.746
	1524.964	-11.9195	12.67927	127.9384
	1904.53	-13.4136	15.83516	141.9849
Mixture (3) [26] **Water (1) + ethane-1,2-diol (2) + propanol (3)**	2800.711	-14.6971	23.28643	190.5616
	2547.764	-14.0756	21.18332	181.0064
	2473.307	-13.9532	20.56424	177.2571
	2389.758	-13.7991	19.86957	173.1819
	2284.342	-13.634	18.9931	167.548
	2101.063	-13.2064	17.46923	159.0939
	1899.338	-12.6844	15.792	149.7385
	1792.776	-12.4537	14.90598	143.955
	1767.944	-12.5135	14.69952	141.2826
	1442.438	-11.6444	11.99311	123.8738
	1904.53	-13.4136	15.83516	141.9849
Mixture (4) [27] **dimethyl carbonate (1) + methanol (2) + ethanol (3)**	1646.216	-12.5991	13.68742	130.6618
	1570.879	-12.479	13.06103	125.8817
	1367.654	-11.911	11.37132	114.823
	1355.522	-11.9292	11.27045	113.6304
	1246.313	-11.634	10.36243	107.127
	1188.008	-11.4738	9.877658	103.5411
	1166.654	-11.4151	9.700114	102.203
	1115.481	-11.2522	9.274638	99.13448
	1489.684	-12.1325	12.38593	122.7849
	1404.552	-11.9836	11.67811	117.2067
	1357.874	-11.8994	11.29001	114.1132
	1280.355	-11.7252	10.64547	109.1968
	1204.133	-11.5213	10.01173	104.5136
	1180.536	-11.4865	9.815533	102.7756
	1300.91	-11.8687	10.81638	109.6088
	1390.66	-11.855	11.5626	117.3059
	1366.565	-11.9022	11.36227	114.8162
	1260.941	-11.6415	10.48406	108.3146
	1003.808	-10.8072	8.346136	92.88361
	1194.154	-11.524	9.928762	103.6229
	1197.942	-11.5482	9.960252	103.7338
	1339.557	-11.7179	11.13771	114.3177
	1327.974	-11.8139	11.0414	112.4075
	1265.691	-11.7006	10.52355	108.173

	1220.328	**-11.61**	**10.14639**	**105.1098**
	1003.392	-10.9026	8.342677	92.03278
	1354.878	-11.8547	11.26509	114.2904
	1299.743	-11.7822	10.80668	110.3146
	1234.66	-11.6424	10.26555	106.0487
	1203.135	-11.5866	10.00343	103.8383
	1327.512	-11.8257	11.03756	112.2569
	1268.538	-11.7302	10.54723	108.1432
	1211.472	-11.6118	10.07275	104.3308
	1292.427	-11.7689	10.74584	109.8176
	1223.924	-11.6355	10.17629	105.1886
	1247.644	-11.6822	10.3735	106.7985
	1255.61	-11.7229	10.43973	107.1072
	1342.972	-11.8601	11.16611	113.2348
	1602.861	-12.3284	13.32695	130.0138
	1150.258	-11.4459	9.563791	100.4957
Mixture (5) [27] **dimethyl carbonate (1) + methanol (2) + hexane (3)**	946.8005	-11.2866	7.872147	83.88703
	921.1461	-11.1775	7.658844	82.41067
	965.0956	-11.2737	8.024261	85.60606
	985.7952	-11.2868	8.196367	87.34039
	1020.092	-11.3256	8.481524	90.06992
	1084.317	-11.4563	9.015525	94.64827
	1034.725	-11.2098	8.603191	92.30546
	1029.805	-11.0807	8.562288	92.93659
	970.7292	-11.3392	8.071101	85.60866
	1013.702	-11.4408	8.4284	88.60445
	1022.184	-11.417	8.498919	89.53178
	1046.111	-11.4357	8.697863	91.47765
	1027.826	-11.3023	8.545834	90.94002
	1070.86	-11.3428	8.903633	94.40882
	1106.645	-11.3658	9.201165	97.3658
	1055.238	-11.5579	8.773745	91.30037
	1061.242	-11.5339	8.823666	92.0107
	1071.722	-11.5209	8.9108	93.02378
	998.9962	-11.1949	8.306127	89.23671
	995.8475	-11.0891	8.279946	89.80396
	1277.691	-11.9522	10.62332	106.9002
	1101.956	-11.6399	9.16218	94.67093
	1129.784	-11.7028	9.393554	96.53932
	1169.733	-11.7658	9.725712	99.41811
	1175.381	-11.729	9.772675	100.2113
	1085.669	-11.333	9.026769	95.79688
	1279.252	-12.1527	10.6363	105.2652
	1151.009	-11.7043	9.570029	98.34057
	1103.259	-11.4889	9.173013	96.02796
	1309.139	-12.0749	10.8848	108.4187
	1205.508	-11.8394	10.02316	101.8221
	1190.300	-11.7697	9.89672	120.2722
	1151.669	-11.5634	9.575518	99.5965
	1189.178	-11.7295	9.887384	101.3839
	1189.16	-11.673	9.887234	101.8723
	1204.732	-11.699	10.01671	102.9778
	1166.805	-11.5007	9.701364	101.455
	1119.444	-11.4538	9.307588	97.73569
	1059.175	-11.5361	8.806479	91.8142
	905.337	-11.1625	7.527399	81.10553

Using the Eq. (9) and Eq. (10), we have estimated the experimental activation energy $(Ea)_{est}$ by replacing the experimental entropic factor $(\ln As)_{exp}$ in Eq. (9). Also, we have estimated the entropic factor $(\ln As)_{est}$ by replacing the experimental activation energy $(Ea)_{exp}$ of ternary liquid mixtures data in Eq. (10). Note that the precedent experimental parameters values $(\ln As)_{exp}$ and $(Ea)_{exp}$ are determined graphically and linear regression, supposing the Arrhenius behavior expressed by Eq. 2 [25, 28-30].

Nevertheless, regarding the five ternary mixtures used in this paper, we note that the fifth mixture, dimethyl carbonate (1), methanol (2) and hexane (3), is a fluid with very low viscosity values. Consequently, in order to take into consideration any eventual effect of such mixture on results [30], our analysis will be made for three different cases separately i.e., using all data, using the first four mixtures of the Table 1, defined as the Group 1 of data, and using the fifth mixture of the Table 1 (dimethyl carbonate (1), methanol (2) and hexane (3)), defined as the Group 2 of data.

Table 2 presents descriptive statistics on experimental and estimated values of parameters for each case i.e, using all data and by groups. The descriptive statistics show that the experimental data are almost similar to the corresponding *Ea*−values and ln*As*−values estimated from Eq. (9) and Eq. (10), particularly for the Group 1. This allows as expecting a good approximation accuracy of the Kacem-Ouerfelli equations for ternary liquids mixture.

Table 2. Descriptive statistics on experimental and estimated values of E_a and ln*As*.

Sample	Size	Parameters	Mean	*σ*	*Min*	*Max*
All data	113	$(E_a)_{exp}$	11.85	3.55	7.53	23.28
		$(E_a)_{est}$	12.32	2.49	8.95	22.05
		$(\ln As)_{exp}$	-12.01	0.78	-14.70	-10.81
		$(\ln As)_{est}$	-11.79	1.13	-14.98	-10.19
Group 1 (4 mixtures)	73	$(E_a)_{exp}$	13.38	3.54	8.34	23.29
		$(E_a)_{est}$	13.18	2.70	8.95	22.05
		$(\ln As)_{exp}$	-12.28	0.82	-14.70	-10.81
		$(\ln As)_{est}$	-12.31	1.08	-14.98	-10.56
Group 2 (Fifth mixture)	40	$(E_a)_{exp}$	9.06	0.82	7.53	10.89
		$(E_a)_{est}$	10.76	0.71	9.63	12.62
		$(\ln As)_{exp}$	-11.50	0.26	-12.15	-11.08
		$(\ln As)_{est}$	-10.85	0.33	-11.56	-10.19

Also, Table 3 presents the Average Absolute Deviations (AAD) for each case, which is a good indicator of quality of approximation [32]. The AAD confirms that the best quality of approximation in mean is clearly in the Group 1 of mixtures where their values is very low for both parameters indicating the little discrepancy between the experimental and the estimated values.

Table 3. The Average Absolute Deviations (AAD %)

Sample	Size	Parameters	
		E_a	ln*As*
All data	113	11.66	3.71
Group 1 (4 mixtures)	73	7.60	2.62
Group 2 (Fifth mixture)	40	19.06	5.70

Table 4. Result of the Wilcoxon Signed-Rank test for both parameters.

Sample	Size	Variables	*z*	Prob> |*z*|
All data	113	$(E_a)_{exp}$ versus $(E_a)_{est}$	-3.24	0.001
		$(lnAs)_{exp}$ versus $(lnAs)_{est}$	-4.24	0.000
Group 1 (4 mixtures)	73	$(E_a)_{exp}$ versus $(E_a)_{est}$	1.48	0.14
		$(lnAs)_{exp}$ versus $(lnAs)_{est}$	1.19	0.23
Group 2 (Fifth mixture)	40	$(E_a)_{exp}$ versus $(E_a)_{est}$	-5.51	0.00
		$(lnAs)_{exp}$ versus $(lnAs)_{est}$	-5.51	0.00

Nevertheless, descriptive statistics are useful but cannot give strong conclusions about the comparison between experimental versus estimated values. For that, there is a need to run a statistical test of comparison of populations such as the Wilcoxon Signed-Rank test [33], 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 statistically identical. Table 4 presents the result of the test for each used sample.

Results of the test confirm the importance of taking into consideration the density of ternary liquid mixtures. Indeed, the Wilcoxon Signed-Rank test leads to reject the null hypothesis for both parameters E_a and ln*As* if all data are used. Also, the null hypothesis is rejected for the group 2, i.e. the mixture of dimethyl carbonate (1), methanol (2) and hexane (3). However, using only the data of Group 1, the null hypothesis becomes accepted indicating that the distributions of the experimental and the estimated values are significantly the same. We deduce from that the validity of the Kacem-Ouerfelli equations for ternary mixture in addition to pure and binary liquid mixtures only for the Group 1.

In order to show the quality of approximation of the Kacem-Ouerfelli equation graphically for ternary mixture of Group 1, we present in Fig. 1 and Fig. 2 the experimental and the estimated values simultaneously for both parameters. Fig. 1 shows the experimental data of one parameter on *x*−axis with experimental and estimated values simultaneously of the second parameter on the *y*−axis. Fig. 2 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, regarding these figures, it is clear that the gap between experimental and estimated values is indicating a slight discrepancy and confirms the good quality of approximation of the Kacem-Ouerfelli equations and its validation for ternary liquid mixtures in addition to pure and binary liquid mixtures for the Group 1 of liquids mixtures.

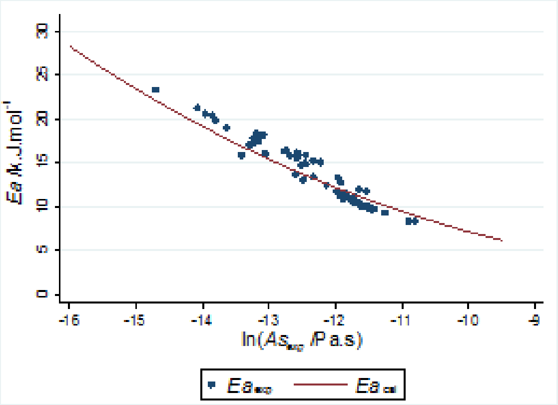

Figure 1. Comparison between the experimental and the estimated values of *Ea* as function of the experimental values $(\ln As)_{exp}$.

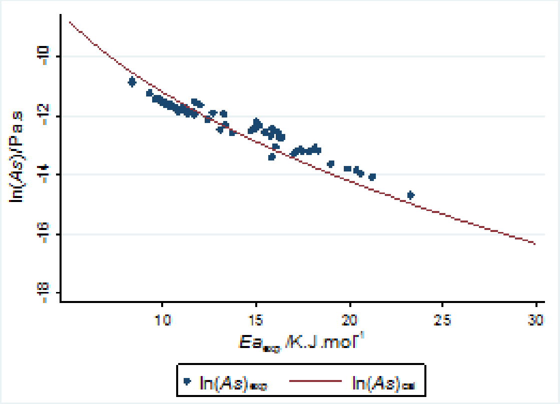

Figure 2. Comparison between the experimental and the estimated values of ln*As* as function of the experimental values $(Ea)_{exp}$.

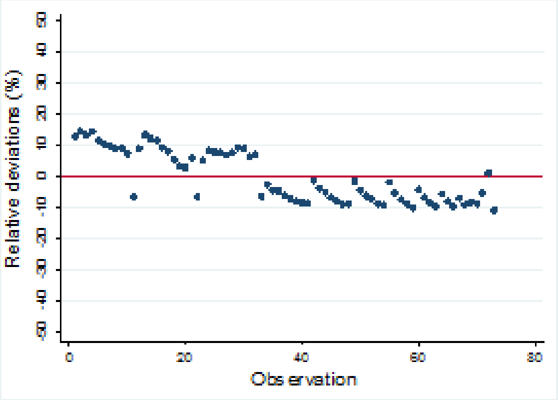

Figure 3 The relative deviations in percentage between experimental and calculated values of *Ea*.

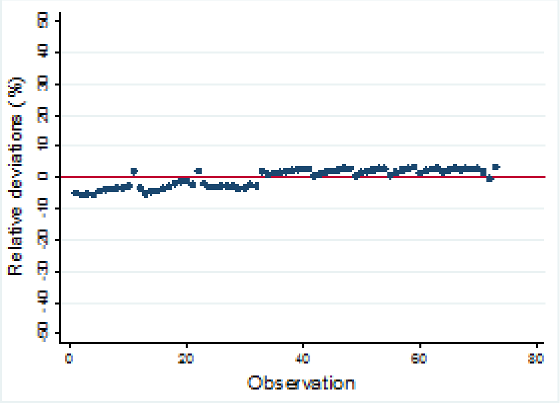

Figure 4 The relative deviations in percentage between experimental and calculated values of ln*As*.

In addition, Fig. 3a and Fig. 3b illustrate the relative deviations in percentage between experimental and calculated values of both parameters for the first group of liquid mixtures. The figures confirm also the precedent result about the good quality of approximation. Indeed, it appears clearly that the relative deviations are very low for both parameters.

Conclusion

Assuming the validity of the equations proposed by Kacem et al. [25] for pure liquids, equations (9) and (10), which model the relationship between the viscosity Arrhenius-type equation parameters, as knowing the entropic factor (ln*As*) and the activation energy (*Ea*), this paper aimed to investigate their validity for ternary liquid mixtures. For that, we used a sample of 114 experimental data from the literature on viscosity for 5 ternary liquid mixtures over different temperature ranges at atmospheric pressure to estimate ln*As* and *Ea*.

In addition, in order to take into consideration, the low viscosity values of the fifth ternary liquid mixture (dimethyl carbonate (1), methanol (2) and hexane (3)), our analysis is made for three different cases i.e., using all data, using the first four mixtures (Group 1 of data), and using the fifth mixture, defined as the Group 2 of data.

At first, a comparison between the experiential values of the parameters *Ea* and ln*As* and their estimated values using Eq. (9) and Eq. (10) is made based on descriptive statistics and the Average Absolute Deviations (AAD). Results show that the experimental data are almost similar to the corresponding estimated values only for the first Group of ternary liquid mixture.

In addition, results of the Wilcoxon Signed-Rank test leads to accept the null hypothesis only for the first group of data indicating that the distributions of the experimental and the estimated values are significantly the same. Different graphical methods are also used; all showed clearly that the gap between experimental and estimated values is indicating a slight discrepancy for the first group of ternary mixture.

We deduced from that the validity of the Kacem-Ouerfelli equations for ternary mixture in addition to pure and binary liquid mixtures only for the first Group of studied liquids mixtures. This indicates also the importance of taking into consideration the density of ternary liquid mixtures in this study.

This result is important in fluids engineering since the validation of these equations for ternary liquid mixtures simplifies the estimation of viscous behavior and the ensuing calculations by reducing the number of viscosity equation parameters and thus facilitating manipulations. We expect that this study will be useful in large domains of applied chemistry and engineering and will open new interesting field of profitable investigations such as the study of specific groups or families of organic liquids solvents. It could also pave the way to estimate more accurate values of the equation's parameters, when the natures of fluids are classified separately.

Disclosure statement

No potential conflict of interest was reported by the authors.

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