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An Extended Belda Equation for Physico-chemical Properties Correlation in Binary Liquid Mixtures at Different Temperatures

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Transport properties, physico-chemical and thermodynamic of fluids are significant for perception of physical phenomena and thermodynamic features and are also required in industrial computations, analytical science, pharmaceutical formulation processes and for scheming and improving manufacturing operations. The majority of such fluid systems manifest non-linear mixtures' behaviour. Consequently rigorous data must be available with semi-empirical equations which are able to furnish a reliable estimation for some physico-chemical quantities of binary liquid systems. The current investigation extends a linking formula supplying a consistent evaluation of physico-chemical features of binary systems of fluids against of mole composition. The validity of the suggested formula has been estimated using well-known binary component solutions and the investigational data of the considered quantities, have been compared with extended equation *versus* Belda one.

Keywords: Binary liquid mixtures, Thermodynamic property, Empirical equation, Correlations, Modeling, Belda equation.

INTRODUCTION

Physico-chemical features of fluid systems are essential for recognizing different thermodynamic behaviours. Diverse physico-chemical characteristics such as density, molar volume, viscosity, surface tension and refractive index should be considered for optimizing and designing fluids [1,2].

Some published articles concerning the physico-chemical quantities of fluid show that the majority of them display non-ideal behaviour or not arranged in a straight line. Accordingly, the estimations of the above quantities or their variations necessitate empirical or semi-empirical formulae able to give a consistent estimation of their specific features [3-5].

The present research suggests an extension of Belda equation [5-7] giving dependable assessment of the special physico-chemical or thermodynamic quantities (density, viscosity, molar volume, surface tension and refractive index, *etc.*) of dual combinations of fluids against of mole composition. The judgment of the investigational data of those quantities *versus* Belda model and our proposed extension has been carried out.

Correlation equations: For most cases mixing operations in applied chemistry and in industrial engineering is difficult due to the non-straight line performance. Then, accurate statistics have to be obtainable with forms able to furnishing a consistent judgment of physical and chemical properties for different behaviours of liquid mixtures. In general, the theoretical description of concepts related to properties is therefore quite complex and exact explicit expressions are absent. This is the reason why several models have been suggested, extended and/or modified over the years referred on some theories, or empirical and semi-empirical equations which are constantly unrelated to every kind of binary liquid mixtures. Moreover, investigations on the physico-chemical quantities of dual mixtures show that a large amount of them reveal non-ideal or non-linear behaviours. Consequently, the search of these quantities or their excess or variations necessitates mathematical expressions eligible of giving a valid approximation of their behaviours [1-8].

In the present work, different properties (Y) were correlated with molar composition using the Belda [5] correlation equation and our proposed extension which uses a correcting factor to the linearity. The equation has been applied at least to 26 binary liquid mixtures. The validity *versus* the magnitude and accuracy of the adjustable parameters is compared and discussed for each property.

A general effective proceeding was used to determinate the deviations ΔY between the experimental values (Y) and the magnitudes which might be gained if the binary systems have been depicted straight line performance, expressed as follows:

$$\Delta Y = Y - (x_1 Y_1 + x_2 Y_2) \quad (1)$$

where Y_1 and Y_2 are the properties of the pure constituent (1) and (2) forming the binary liquid mixture at given molar fraction (x_1, x_2).

Thus, these differences are subjected to non-linear regression correlation according to the famous polynomial Redlich-Kister equation [3]:

$$\Delta Y = Y^E = x_1 \cdot (1 - x_1) \sum_{p=0}^{p=n} A_{n,p,T} \cdot (2x_1 - 1)^p \quad (2)$$

where x_1 is the molar composition of pure liquid constituent (1) and $A_{n,p,T}$ the adjustable coefficient for every degree p of one-term expression $(2x_1 - 1)^p$ at absolute temperature T and,

$$\Delta Y = Y^E = x_1 \cdot (1 - x_1) \cdot Q_{RK,Y}(x_1) \quad (3)$$

where the reduced Redlich-Kister property $Q_{RK,Y}(x_1)$ is expressed as follows [9-20]:

$$Q_{RK,Y}(x_1) = \sum_{p=0}^{p=n} A_{n,p,T} \cdot (2x_1 - 1)^p = A_o + A_1 \cdot (2x_1 - 1) \quad (4)$$

Belda equation: Generally, the proposed correlation equations are based on the deviation on the linear or ideal behaviour ($Y_{mixture} = \Sigma_{i=1}^{n} Y_i x_i$) of binary liquid mixtures expressed as follows:

$$Y(x) = Y_2 + (Y_1 - Y_2) \cdot x_1 \quad (5)$$

So, Belda [5] proposed an empirical correlation equation (eqn. 6) with two adjustable parameters (m_1, m_2) for investigated four physico-chemical properties such as (density, viscosity, molar volume and refractive index) which introduces a correcting factor $F_{B,Y}(x_1)$ (*i.e.* fraction containing the two parameters) as an homographic function (eqn. 7) depending on the mole composition of one pure component of binary liquid system (x_1).

$$Y(x) = Y_2 + (Y_1 - Y_2) \cdot x_1 \cdot \frac{1 + m_1 \cdot (1 - x_1)}{1 + m_2 \cdot (1 - x_1)} \quad (6)$$

where Y_1 is the larger magnitudes of the pure liquid constituents constituting the binary system with molar composition (x_1, x_2) and m_1, m_2 are two adjustable parameters to be fitted.

$$F_{B,Y}(x_1) = \frac{1 + m_1 \cdot (1 - x_1)}{1 + m_2 \cdot (1 - x_1)} \quad (7)$$

It is known that the Belda equation responds to straight line attitude when $m_1 = m_2$, *i.e.* while the correction operator $F_{B,Y}(x_1) = 1$. In the case, where the correcting operator is > 1, positive synergy of the quantity has been recorded, whereas $F_{B,Y}(x_1) < 1$ such synergy proves negative. We add that more the difference between the Belda parameters are accentuated, more the system behaviour deviates to the linearity.

It is noticed that the Belda model generally success for numerous binary mixtures. Nevertheless, for some particular situations, we observe small discrepancy [6,7]. In fact, due to the few adjustable parameters (m_1 and m_2) and the homographic expression of $F_{B,Y}(x_1)$ which doesn't present any change of curvature (inflection point), Belda equation exhibits low performance when the curves representing the binary mixture property (Y) indicates some peculiarities (inflection points, abrupt change of curvature, *etc.*) due to the existence of some strong correlation at particular compositions [6,7]. So, we observe that some small discrepancies between experimental data points and the fitted model, especially at the boundary limit of the composition domain and when the Belda factor exhibit a vertical asymptote very near or into the interval of composition (Figs. 1 and 2).

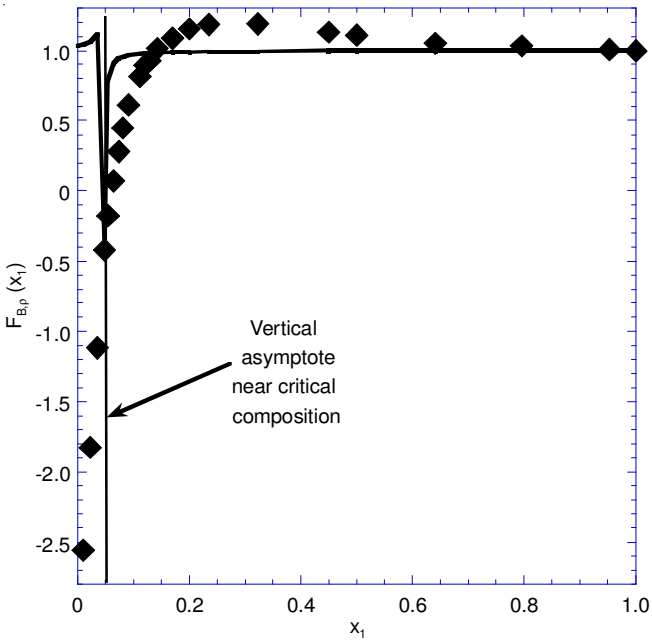

Fig. 1. Variation of Belda factor for the density $F_{B,r}(x_1)$ for isobutyric acid (1) + water (2) systems against molar fraction x_1 at 308.15 K (◆): experimental, (—): calculated by (eqn. 6) [Ref. 6]

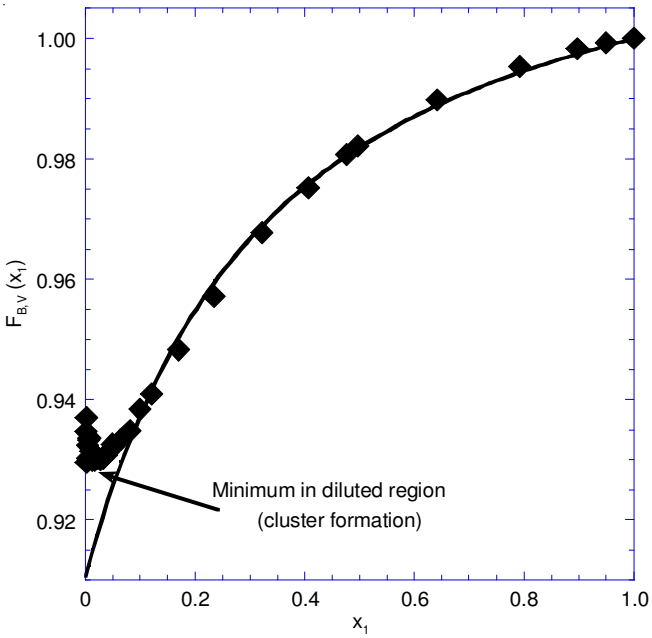

Fig. 2. Variation of Belda factor for the molar volume $F_{B,V}(x_1)$ for 1,4-dioxanne (1) + water (2) systems against molar fraction x_1 at 298.15 K. (◆): experimental, (—): calculated by (eqn. 6) [Ref. 7]

Proposed extended Belda equation: For the precedent reasons we understand about an addition of only one new parameter (m_3) on the Belda model. Then, following the precedent mathematical analysis, we propose a supplementary adjustable m_3-parameter associated to the mole fraction x_2 square and added on the numerator of the Belda factor which can be expressed as follows:

$$Y(x) = Y_2 + (Y_1 - Y_2)\cdot x_1 \cdot \frac{1 + m_1\cdot(1-x_1) + m_3\cdot(1-x_1)^2}{1 + m_2\cdot(1-x_1)} \quad (8)$$

where Y_1 is the larger of the magnitudes of the pure liquid constituents constituting the binary system with molar composition (x_1, x_2) and m_1, m_2 and m_3 are three adjustable parameters to be fitted.

RESULTS AND DISCUSSION

To examine the suitability of the proposed correlation equation, 26 published systems in literature have been utilized [5,21-27] to investigate the density, viscosity and molar volume (Table-1) and 33 systems for refractive index (Table-2) of diverse dual liquids systems at 1 atmosphere with dissimilar temperature values. The results of these samples magnitudes have been compared with those achieved by the suggested formula, whereas Belda mathematical form arranging as comparing factor for the standard deviation σ assigned as:

$$\sigma = \sqrt{\frac{\sum_{i=1}^{i-N}(Y_{i,exp} - Y_{i,cal})^2}{N - K}} \quad (9)$$

TABLE-1
CONSTITUENTS OF THE MIXTURES UTILIZED IN THE REFERENCE SYSTEMS FOR FITTING DENSITY, VISCOSITY AND MOLAR VOLUME (N IS THE NUMBER OF EXPERIMENTAL DATA POINTS)

System	T (K)	Binary mixture		N	Ref.
		Component (1)	Component (2)		
1	303.15	1,3-Dioxolane	Methanol	11	[21]
2			Ethanol		
3			1-Propanol		
4			2-Propanol		
5			1-Butanol		
6			2-Butanol		
7			*t*-Butanol		
8			i-Amyl alcohol		
9	293.15	2-Propanol	Octane	12	[22]
10	298.15				
11	303.15				
12	293.15	2-Propanol	Decane		
13	298.15				
14	303.15				
15	293.15	2-Propanol	Dodecane		
16	298.15				
17	303.15				
18	293.15	2-Butanol	Octane	13	[23]
19	298.15				
20	303.15				
21	293.15	2-Butanol	Decane		
22	298.15				
23	303.15				
24	293.15	2-Butanol	Dodecane		
25	298.15				
26	303.15				

TABLE-2
CONSTITUENTS OF THE MIXTURES UTILIZED IN THE REFERENCE SYSTEMS FOR FITTING REFRACTIVE INDEX (N IS THE NUMBER OF EXPERIMENTAL DATA POINTS)

System	T (K)	Binary mixture		N	Ref.
		Component (1)	Component (2)		
1	298.15	Methyl-cyclohexane	Ethanol	11	[24]
2	303.15				
3	308.15				
4	298.15		Propan-1-ol		
5	303.15				
6	308.15				
7	298.15		Propan-2-ol		
8	303.15				
9	308.15				
10	298.15		Butan-1-ol		
11	303.15				
12	308.15				
13	298.15		2-Methyl-1-propanol		
14	303.15				
15	308.15				
16	298.15		3-Methyl-1-butanol		
17	303.15				
18	308.15				
19	303.15	1-Butanol	2-Butanone	17	[25]
20	300.15	Isobutyric acid	Water		[26]
21	303.15				
22	308.15				
23	313.15				
24	293.15	1,1,2,2-Tetrabromoethane	1-Pentanol	11	[27]
25	303.15				
26	298.15		1-Hexanol		
27	303.15				
28	298.15		1-Heptanol		
29	303.15				
30	293.15		1-Octanol		
31	303.15				
32	293.15		1-Dectanol		
33	303.15				

whereas N is the amount of outcome points of the sample and k is the amount of free adjustable factors of the proper equations 6 and 8 (N = k+1). The most selective factor m-values (eqn. 8) and the corresponding standard deviation σ are given in Tables 3-6 for each physico-chemical property, separately. We have treated the same systems chosen by Belda to estimate the goodness of the suggested extension. The results of this study have been compared with those obtained with the Belda correlation model [4-9]. Figs. 3-6 show the variation of the obtained standard deviations values.

Tables 3-6 report the amounts of the optimal m_i-parameters calculated through non-linear regression of the eqn. 8 and the corresponding standard deviations σ, the mean standard deviation $\overline{\sigma}$ and limiting standard deviations σ_{min} and σ_{max} of each set of the studied properties mentioned in Tables 1 and 2.

The obtained statistical data are also illustrated in Figs. 3-6. It is observed that globally the standard deviations (σ)-values of the extended model (eqn. 8) are less than those of the Belda model (eqn. 6) especially for high values of standard deviations related to Belda model. Nevertheless, it is noticed that for some rare cases for low standard deviation values, Belda

TABLE-3
OPTIMAL COEFFICIENTS m_i FOR eqn. 8, STANDARD DEVIATIONS σ CALCULATED BY eqn. 9, MEAN STANDARD DEVIATIONS $\bar{\sigma}$ AND STANDARD DEVIATIONS σ_{min} AND σ_{max} OF DENSITY, FOR THE INDICATED REFERENCE SYSTEMS (TABLE-1)

System	Belda eqn. 6	Extended Belda eqn. 8			
	σ	m_1	m_2	m_3	σ
1	0.00025	0.22846	-0.23037	0.67106	0.00010
2	0.00003	0.02683	-0.14071	0.49748	0.00002
3	0.00014	-1.10998	-1.01455	-0.0877	0.00012
4	0.00221	-1.10201	-0.96883	-0.11746	0.00007
5	0.00014	-10.31184	-9.99356	-0.30841	0.00006
6	0.00034	-10.26888	-9.92402	3.41845	0.00005
7	0.00017	-10.42721	-10.05528	3.77761	0.00006
8	0.00013	-12.06307	-11.57409	-5.64573	0.00007
9	0.00027	0.60404	2.04507	-0.73413	0.00008
10	0.00031	0.80531	2.30799	-0.88246	0.00006
11	0.00042	0.87914	2.39704	-1.00526	0.00009
12	0.00026	0.46471	2.618	-0.81826	0.00010
13	0.00031	0.57953	2.79206	-1.00629	0.00012
14	0.00031	0.46675	2.68989	-0.96573	0.00013
15	0.00023	0.59451	3.64406	-1.36817	0.00004
16	0.00025	0.54694	3.6472	-1.42726	0.00006
17	0.00028	0.53436	3.69932	-1.59043	0.00008
18	0.00018	0.16518	1.07502	-0.27434	0.00009
19	0.00018	0.09931	1.02845	-0.2261	0.00010
20	0.00018	0.18312	1.1262	-0.30205	0.00011
21	0.00019	0.13369	1.51962	-0.36949	0.00010
22	0.00019	0.18702	1.60066	-0.42525	0.00008
23	0.00022	0.23819	1.68697	-0.47968	0.00010
24	0.00007	-0.43093	1.40527	0.08303	0.00004
25	0.00009	-0.24674	1.66447	-0.09341	0.00005
26	0.00010	-0.27215	1.67824	-0.10416	0.00006
$\bar{\sigma}$	0.00028			$\bar{\sigma}$	0.00016
σ_{max}	0.00221			σ_{max}	0.00073
σ_{min}	0.00003			σ_{min}	0.00002

TABLE-4
OPTIMAL COEFFICIENTS m_i FOR eqn. 8, STANDARD DEVIATIONS σ CALCULATED BY eqn. 9, MEAN STANDARD DEVIATIONS $\bar{\sigma}$ AND STANDARD DEVIATIONS σ_{min} AND σ_{max} OF VISCOSITY, FOR THE INDICATED REFERENCE SYSTEMS (TABLE-1)

System	Belda eqn. 6	Extended Belda eqn. 8			
	σ	m_1	m_2	m_3	σ
1	0.01484	1.36219	-1.00529	-1.8562	0.00706
2	0.02376	2.3251	2.42218	2.84229	0.01005
3	0.02041	0.1882	-0.93616	-3.49234	0.01205
4	0.01567	0.11808	-0.79329	1.25602	0.00850
5	0.01348	0.03874	-0.86603	-4.93178	0.00844
6	0.01091	0.10266	-0.85808	-0.72532	0.01041
7	0.01359	0.17931	-0.83156	-0.43881	0.01525
8	0.02066	0.18103	-0.47605	2.61678	0.01402
9	0.00640	-0.72721	3.97017	0.38647	0.00802
10	0.00396	-0.70928	3.87451	0.47134	0.00257
11	0.00503	-0.89482	3.4826	0.11542	0.00560
12	0.00851	-0.15982	5.18351	10.36707	0.00469
13	0.00836	-0.40887	4.83438	3.26952	0.00660
14	0.00854	-0.50873	4.76065	2.74093	0.00555
15	0.01026	0.24296	7.52527	-26.4459	0.00544
16	0.00867	-0.59082	7.06296	9.99937	0.00464
17	0.00775	-1.65947	6.27164	3.00717	0.00558
18	0.00896	-1.28052	4.05704	-0.42737	0.00422
19	0.00756	-1.21926	3.90486	-0.40462	0.00551
20	0.00756	-1.28773	3.50846	-0.44031	0.00708
21	0.00420	-0.98762	4.85417	0.17487	0.00351
22	0.00561	-0.96701	4.72777	0.24787	0.00475
23	0.00635	-1.00589	4.48031	0.24752	0.00492
24	0.00358	-1.52369	4.76105	0.13384	0.00580
25	0.00280	-1.68283	4.58094	0.09496	0.00395
26	0.00318	-1.7455	4.50224	0.22966	0.00367
$\bar{\sigma}$	0.00963			$\bar{\sigma}$	0.00684
σ_{max}	0.02376			σ_{max}	0.01525
σ_{min}	0.00280			σ_{min}	0.00257

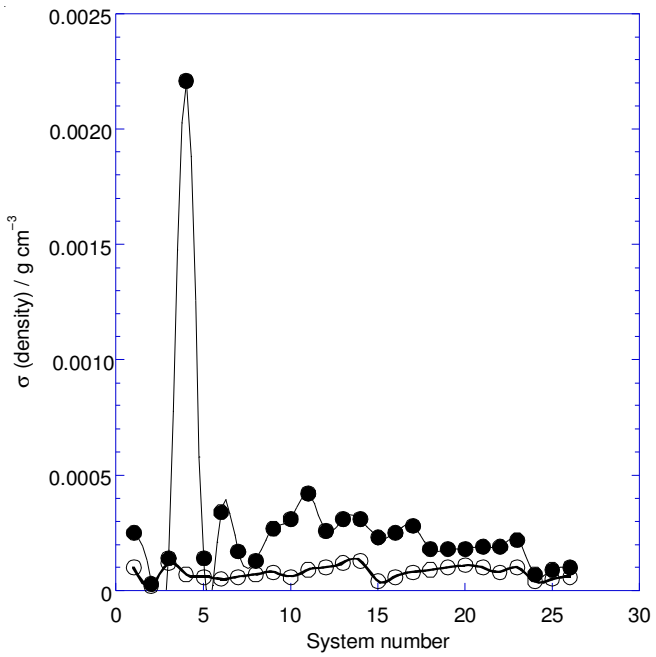

Fig. 3. Comparison of standard deviations σ for density (Table-3) calculated by eqn. (9), related to (•): Belda model (eqn. 6) and that (o): calculated by proposed equation (eqn. 8)

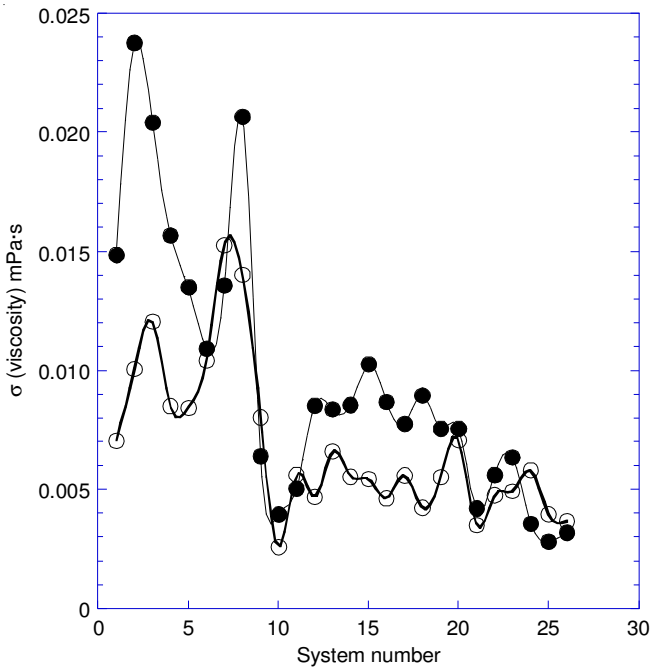

Fig. 4. Comparison of standard deviations σ for viscosity (Table-4) calculated by eqn. (9), related to (•): Belda model (eqn. 6) and that (o): calculated by proposed equation (eqn. 8)

TABLE-5
OPTIMAL COEFFICIENTS m_i FOR eqn. 8, STANDARD DEVIATIONS σ CALCULATED BY eqn. 9, MEAN STANDARD DEVIATIONS $\bar{\sigma}$ AND STANDARD DEVIATIONS σ_{min} AND σ_{max} OF MOLAR VOLUME, FOR THE INDICATED REFERENCE SYSTEMS (TABLE-1)

System	Belda eqn. 6	Extended Belda eqn. 8			
	σ	m_1	m_2	m_3	σ
1	0.01007	-0.86177	-0.82352	0.02933	0.01249
2	0.00219	-0.23076	-0.22818	-0.01262	0.00168
3	0.00976	-1.16386	-1.06505	0.10453	0.00978
4	0.00678	0.20276	0.28921	-0.12268	0.00581
5	0.00968	-0.61235	-0.57635	0.00956	0.00956
6	0.01899	-0.85784	-0.80166	0.02664	0.01690
7	0.00993	-0.83618	-0.77177	0.03772	0.00726
8	0.00997	-0.72301	-0.69847	0.01270	0.00967
9	0.01854	-0.89685	-0.87287	0.01687	0.01031
10	0.02068	-0.91631	-0.88988	0.02022	0.01312
11	0.02503	-0.94202	-091381	0.02190	0.01239
12	0.03323	-1.03641	-1.01628	0.01992	0.02192
13	0.03833	-1.03207	-1.01026	0.02124	0.02394
14	0.03972	-1.03435	-1.01110	0.02262	0.01931
15	0.02879	-1.01002	-0.99408	0.01534	0.02156
16	0.03253	-1.01816	-1.00119	0.01642	0.02140
17	0.03647	-1.01613	-0.99784	0.01746	0.01948
18	0.01810	-0.88716	-0.86130	0.01646	0.01421
19	0.02042	-0.94088	-0.91173	0.02255	0.01535
20	0.02361	-0.94385	-0.91256	0.02374	0.01629
21	0.02293	-0.96499	-0.94268	0.01881	0.01588
22	0.02101	-0.94934	-0.92567	0.01938	0.01114
23	0.02581	-0.98479	-0.95900	0.02257	0.00793
24	0.00780	-1.01809	-0.99970	0.01846	0.00772
25	0.01235	-1.04277	-1.02310	0.02004	0.01104
26	0.01532	-1.05219	-1.03082	0.02191	0.01166
$\bar{\sigma}$	0.01992			$\bar{\sigma}$	0.01337
σ_{max}	0.03972			σ_{max}	0.02394
σ_{min}	0.00219			σ_{min}	0.00168

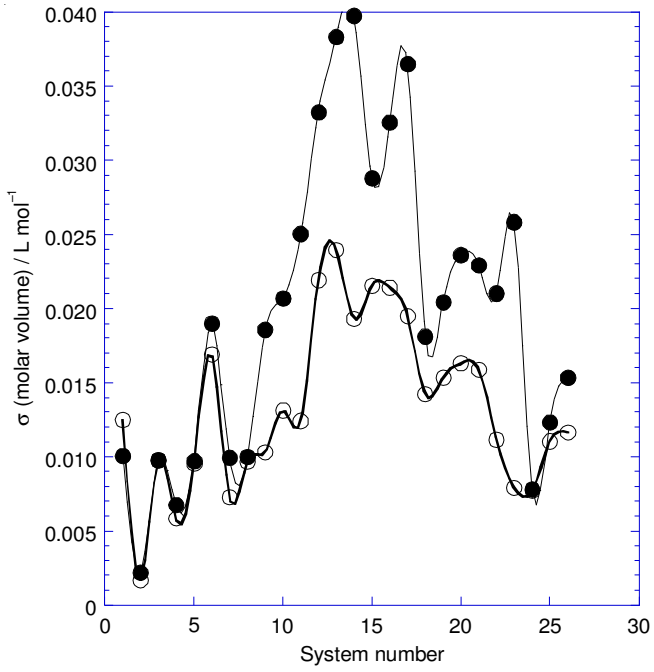

Fig. 5. Comparison of standard deviations σ for molar volume (Table-5) calculated by eqn. (9), related to (•): Belda model (eqn. 6) and that (o): calculated by proposed equation (eqn. 8)

TABLE-6
OPTIMAL COEFFICIENTS m_i FOR eqn. 8, STANDARD DEVIATIONS σ CALCULATED BY eqn. 9, MEAN STANDARD DEVIATIONS $\bar{\sigma}$ AND STANDARD DEVIATIONS σ_{min} AND σ_{max} OF REFRACTIVE INDEX, FOR THE INDICATED REFERENCE SYSTEMS (TABLE-2)

System	Belda eqn. 6	Extended Belda eqn. 8			
	$\sigma \times 10^4$	m_1	m_2	m_3	$\sigma \times 10^4$
1	3.4962	-4.56945	-5.22075	-3.42032	1.9096
2	3.0039	-4.26530	-4.87565	-2.96848	2.8978
3	3.9007	-9.48039	-10.08974	-6.10193	2.7843
4	1.5806	-9.71669	-9.99014	-2.73488	1.4246
5	5.4645	-1.00342	-1.00221	-0.36129	0.57615
6	1.8819	-0.72675	-1.02879	-0.32537	1.1122
7	1.7858	2.18506	1.99638	0.83762	1.5489
8	1.3777	1.47670	1.30540	0.58094	1.1199
9	0.72701	-0.20866	-0.44003	-0.02779	0.72019
10	3.1571	-1.08189	-1.09335	0.01479	3.1411
11	2.9737	-1.07667	-1.10266	-0.01229	2.9425
12	2.9293	-1.10935	-1.11193	-0.00016	2.8906
13	0.95528	-0.83679	-0.96778	-0.09332	0.75305
14	0.77863	-4.15369	-4.21843	-0.28584	0.71885
15	1.8280	-0.97802	-1.04913	-0.08626	1.3497
16	0.13622	-0.97410	-0.97189	0.01060	0.06333
17	0.80877	-4.59138	-4.56172	0.11873	0.7368
18	0.7016	-1.00596	-0.99870	0.00804	0.08323
19	0.30001	1.80555	1.39743	-0.17674	0.2275
20	2.1320	1.95008	0.88942	-0.16919	2.1097
21	2.1299	1.45387	0.43868	-0.34044	1.9465
22	2.4139	1.15686	0.15363	-0.49812	2.022
23	2.4374	1.14688	0.12585	-0.53220	1.9645
24	0.54009	-1.00785	-0.99932	0.00891	0.16945
25	0.43871	-1.01182	-0.99921	0.01304	0.60942
26	0.47931	0.23815	0.40108	-0.01976	0.48172
27	0.47913	-0.57209	-0.40441	0.08782	0.33146
28	0.37215	-0.11803	0.19075	0.04539	0.29141
29	0.22309	-0.13033	0.18416	0.03581	0.1681
30	0.47743	0.23598	0.69500	-0.07558	0.36709
31	0.48552	-0.08898	0.37474	0.03316	0.44517
32	0.4558	0.02417	0.77835	0.00036	0.4558
33	0.3100	-0.00682	0.76251	0.00438	0.29926
$\bar{\sigma}\times10^4$	1.55034			$\bar{\sigma}\times10^4$	1.17157
$\sigma_{max}\times10^4$	5.4645			$\sigma_{max}\times10^4$	3.14110
$\sigma_{min}\times10^4$	0.13622			$\sigma_{min}\times10^4$	0.06333

model excels. This can be explained in the case of monotonous variation of the studied properties against molar composition, the two parameter model is optimal, while the statistical analysis of a model with more than two adjustable parameters can give absurd parameters values or doesn't well converge in the iterations calculation by the used software. So, when there are no special phenomena occurring in the binary system, we prefer the Belda model than our extended model. Nevertheless, when there are strong curvatures, singular points or change of curvature, our extended models provided better results. Figs. 7-10 show how the excellent agreement between experimental data points and the calculated values of physico-chemical properties in the whole range of molar compositions of the some studied binary liquid mixtures.

Physical meaning of Belda parameters: Comparing the Redlich-Kister expression (eqns. 2-4) and the Belda model (eqn. 6), we can deduce the following relationship:

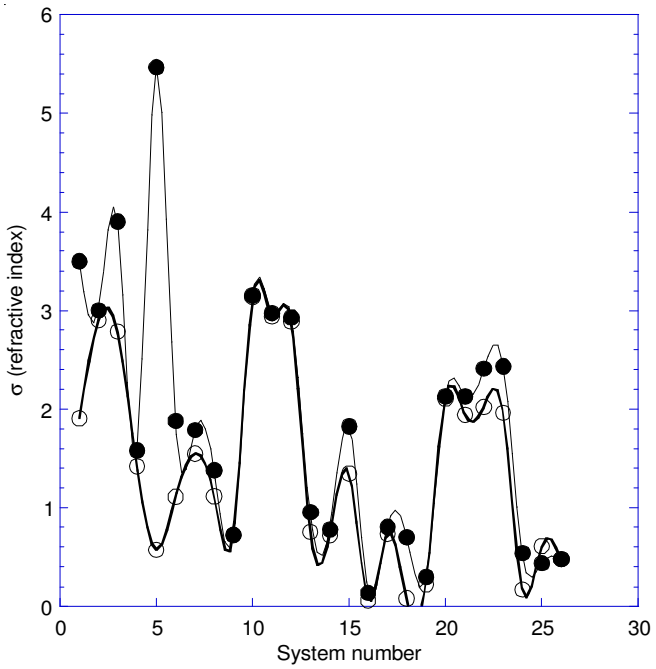

Fig. 6. Comparison of standard deviations σ for refractive index (Table-6) calculated by eqn. (9), related to (•): Belda model (eqn. 6) and that (o): calculated by proposed equation (eqn. 8)

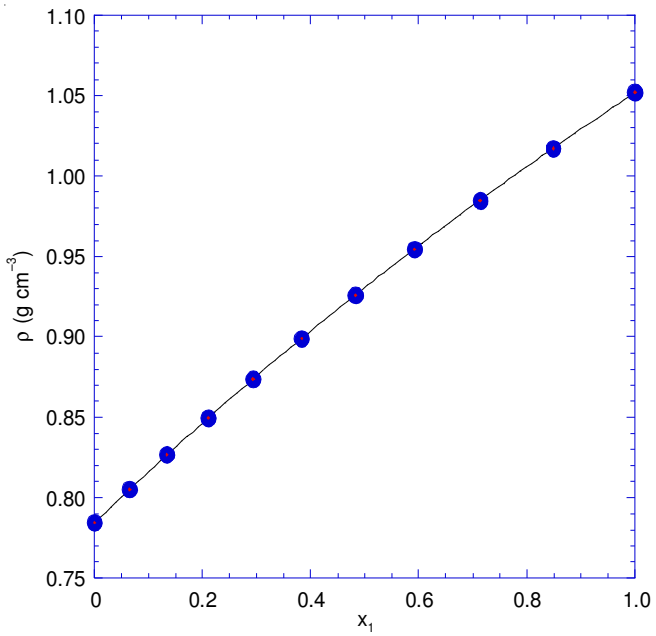

Fig. 7. Comparison of experimental density data at with that calculated by proposed equation (eqn. 8) (system 2, Tables 1 and 3)

$$Q_{RK,Y}(x_1) = \frac{Y_1 - Y_2}{1 + m_2 \cdot (1 - x_1)}[(m_1 - m_2) + m_3(1 - x_1)] \quad (10)$$

In relation with previous works [9-20] we have demonstrated the correlation among the reduced Redlich-Kister function $Q_{RK,Y}(x_1)$ and the apparent molar thermodynamic quantities ($Y_{i,\phi}$) for one component (i) as follows:

$$Q_{RK,Y}(x_1) = \frac{Y^E}{x_1(1-x_1)} = \frac{Y_{1,\varphi} - Y_1^*}{1 - x_1} = \frac{Y_{2,\varphi} - Y_2^*}{x_1} \quad (11)$$

We can write then:

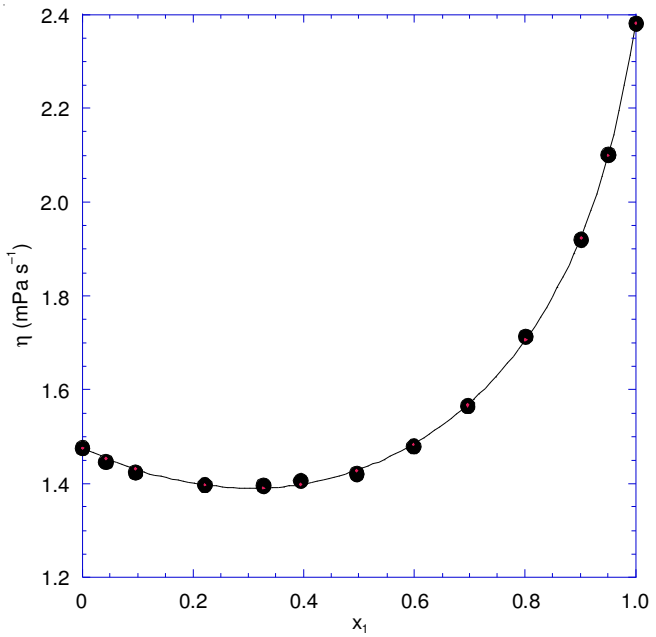

Fig. 8. Comparison of experimental viscosity data at with that calculated by proposed equation (eqn. 8) (system 15, Tables 1 and 4)

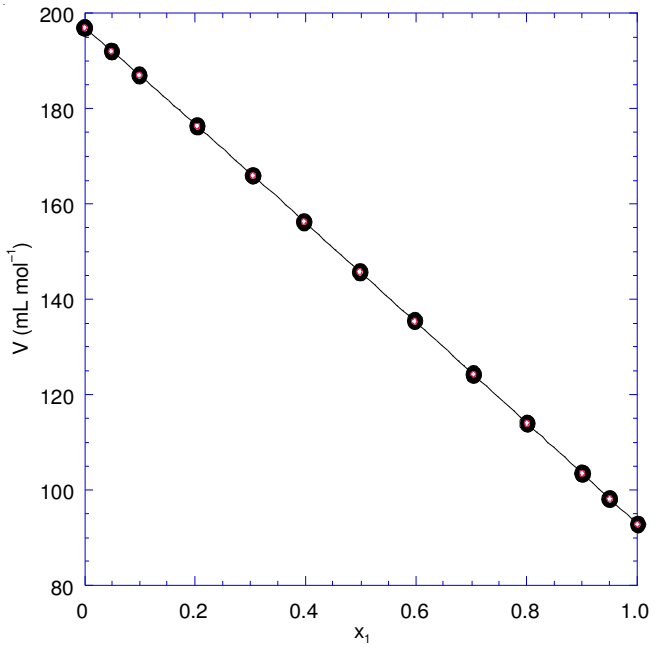

Fig. 9. Comparison of experimental volume data at with that calculated by proposed equation (eqn.8) (system 23, Tables 1 and 4)

$$Y_{1,\varphi}(x_1) = Y_1 + x_2 Q_{RK,Y}(x_1) \quad (12)$$

$$Y_{2,\varphi}(x_1) = Y_2 + x_1 \quad (13)$$

Replacing eqn. 10 into eqns. 12 and 13, we obtain:

$$Y_{1,\varphi}(x_1) = Y_1 + \frac{(Y_1 - Y_2)\cdot x_2}{1 + m_2 \cdot (1 - x_1)} \cdot [(m_1 - m_2) + m_3 \cdot (1 - x_1)] \quad (14)$$

$$Y_{2,\varphi}(x_1) = Y_2 + \frac{(Y_1 - Y_2)\cdot x_2}{1 + m_2 \cdot (1 - x_1)} \cdot [(m_1 - m_2) + m_3 \cdot (1 - x_1)] \quad (15)$$

Likewise, we can obtain the limiting excess partial molar thermodynamic quantities ($\overline{Y}_i^{E,\infty}$) as follow:

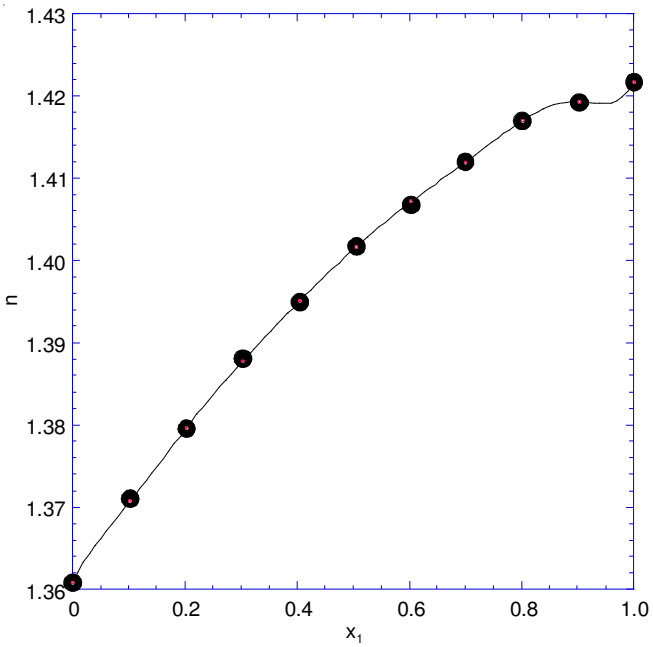

Fig. 10. Comparison of experimental refractive index data at with that calculated by proposed equation (eqn. 8) (system 1, Tables 2 and 6)

$$\bar{Y}_1^{E,\infty} = Q_{RK,Y}(x_1 = 0) = \bar{Y}_1^{\infty} - Y_1 = \frac{m_1 - m_2 - m_3}{1 + m_2}(Y_1 - Y_2) \quad (16)$$

and,

$$\bar{Y}_2^{E,\infty} = Q_{RK,Y}(x_1 = 0) = \bar{Y}_2^{\infty} - Y_2 = (m_1 - m_2)(Y_1 - Y_2) \quad (17)$$

So, it is concluded that the obtained values of these thermodynamic quantities simply using the adjustable m_i-parameters values after fitting the correlation between experimental data points and the Belda model or our extended suggested equation. Moreover, in the case of Belda model (m_1, m_2) it is concluded that (m_2)-parameter represents a relative deviation between the two partial molar quantities:

$$m_2 = \frac{(\bar{Y}_1 - \bar{Y}_2)}{\bar{Y}_1} \quad (18)$$

while the (m_1)-parameter is expressed as follow:

$$m_1 = \frac{\bar{Y}_2}{Y_1 - Y_2} + m_2 \quad (19)$$

In case of the present suggested extended model, the presence of the third (m_3)-parameter complicates the mathematical manipulations and requires another independent equation to solve a system of three equations and gives an expression for each m_i-parameter in relationship with classical thermodynamic quantity.

Conclusions

To examine the suitability of the proposed correlation equation, 26 published systems in the literature have been exploited investigating the density, viscosity and molar volume and 33 systems for refractive index of diverse dual liquids combinations have been explored at 1 atmosphere with dissimilar temperature values. The results of these samples have been compared with those achieved by the suggested formula, whereas Belda mathematical form arranging as comparing factor for the standard deviation σ.

We have treated the same systems chosen by Belda due to evaluate the goodness of the suggested extension. Using the two different expressions of Belda model (the original one with two parameters and our suggested extended equation using three adjustable parameters) in 26 or 33 samples of binary mixtures identified indicated above and comparing the standard deviation, we see that the proposed model offer better results than those of Belda especially for high standard deviation.

We have also concluded that due to the few adjustable Belda parameters (m_1 and m_2) and the homographic shape of the correcting Belda factor which doesn't present any change of curvature (inflection point). Belda equation exhibited low performance when the curves representing the binary mixture property indicating some peculiarities (inflection points, abrupt change of curvature, *etc.*) due to the existence of some strong correlation at particular compositions. So, we have observed some small discrepancies between experimental data points and the fitted model, especially at the high dilution and when the Belda factor exhibited a vertical asymptote very near or into the range of composition. Nevertheless, when there are strong curvatures, singular points or change of curvature, our extended models gives better results. Excellent agreement has been obtained between experimental data points and the calculated values of physico-chemical properties in the whole range of molar compositions of the some studied binary liquid mixtures. Due the obtained improvements, we can ascertain that the suggested extended model can be used to correlate several physico-chemical properties in binary liquid mixtures, more than the fourth properties tested by Belda.

We have demonstrated that the adjustable parameters have some physical significance and they are in relationship with the apparent molar thermodynamic quantities and the limiting excess partial molar thermodynamic quantities. As a result, the Belda model can be upgraded and considered as a semi-empirical equation and can be used as new techniques to determinate these thermodynamic quantities with a reliable estimation. The Belda model and our extended suggested equation can be used with reliable correlation for any physico-chemical properties other tested by Belda. In future works, we will study about the supplementary third adjustable parameter which can be added in the denominator of the correcting Belda factor and compare the goodness of different models.

REFERENCES

1. H.D. Crockford and B. Knight, Fundamentos de Fisico-Química, C.E.C.S.A., Madrid (1968).
2. S. Glasstone, Tratado de Química Física, Aguilar, Madrid (1970).
3. O. Redlich and A.T. Kister, *Ind. Eng. Chem.*, **40**, 345 (1948); https://doi.org/10.1021/ie50458a036.
4. R.C. Reid, J.M. Prausnitz and T.M. Sherwood, The Properties of Gases and Liquids, MacGraw Hill, New York (1977).
5. R. Belda, *Fluid Phase Equilib*., **282**, 88 (2009) https://doi.org/10.1016/j.fluid.2009.04.023.
6. N. Ouerfelli, A. Messâadi, E.B.H. H'mida, E. Cherif and N. Amdouni, *Phys. Chem. Liq.*, **49**, 655 (2011); https://doi.org/10.1080/00319104.2010.517204.
7. N. Ouerfelli, O. Iulian, R. Besbes, Z. Barhoumi and N. Amdouni, *Phys. Chem. Liq.*, **50**, 54 (2012); https://doi.org/10.1080/00319104.2010.494246.
8. A. Messaâdi, N. Ouerfelli, D. Das, H. Hamda and A.H. Hamzaoui, *J. Solution Chem.*, **41**, 2186 (2012); https://doi.org/10.1007/s10953-012-9931-3.

9. J.E. Desnoyers and G. Perron, *J. Solution Chem.*, **26**, 749 (1997); https://doi.org/10.1007/BF02767781.
10. H. Salhi, S. Babu, N. Al-Eidan, N.H. Mekni, N. Al-Otaibi, K.Y. Alqahtani, N.A. Al-Omair and N. Ouerfelli, *Mediterr. J. Chem.*, **6**, 33 (2017); https://doi.org/10.13171/mjc62/01701061439-salhi.
11. H. Salhi, S. Babu, A.A. Al-Arfaj, M.A. Alkhaldi, N.O. Alzamel, S. Akhtar and N. Ouerfelli, *Rasayan J. Chem.*, **9**, 864 (2016).
12. N. Ouerfelli, Z. Barhoumi, R. Besbes and N. Amdouni, *Phys. Chem. Liq.*, **49**, 777 (2011); https://doi.org/10.1080/00319104.2010.521927.
13. D. Das, Z. Barhoumi and N. Ouerfelli, *Phys. Chem. Liquids*, **50**, 346 (2012); https://doi.org/10.1080/00319104.2011.646516.
14. D. Das and N. Ouerfelli, *J. Solution Chem.*, **41**, 1334 (2012); https://doi.org/10.1007/s10953-012-9878-4.
15. D. Das, A. Messaâdi, Z. Barhoumi and N. Ouerfelli, *J. Solution Chem.*, **41**, 1555 (2012); https://doi.org/10.1007/s10953-012-9888-2.
16. D. Das, Z. Barhoumi, N. Dhouibi, M.A.M.K. Sanhoury and N. Ouerfelli, *Phys. Chem. Liq.*, **50**, 712 (2012); https://doi.org/10.1080/00319104.2012.713553.
17. D. Das, A. Messaâdi, N. Dhouibi and N. Ouerfelli, *Phys. Chem. Liq.*, **50**, 773 (2012); https://doi.org/10.1080/00319104.2012.717893.
18. N. Ouerfelli, T. Kouissi, N. Zrelli and M. Bouanz, *J. Solution Chem.*, **38**, 983 (2009); https://doi.org/10.1007/s10953-009-9423-2.
19. N. Ouerfelli, T. Kouissi and O. Iulian, *J. Solution Chem.*, **39**, 57 (2010); https://doi.org/10.1007/s10953-009-9484-2.
20. N. Ouerfelli, O. Iulian and M. Bouaziz, *Phys. Chem. Liq.*, **48**, 488 (2010); https://doi.org/10.1080/00319100903131559.
21. M.N. Roy, A. Sinha and B. Sinha, *J. Solution Chem.*, **34**, 1311 (2005); https://doi.org/10.1007/s10953-005-8022-0.
22. B. González, A. Domínguez and J. Tojo, *J. Chem. Thermodyn.*, **35**, 939 (2003); https://doi.org/10.1016/S0021-9614(03)00047-8.
23. B. González, A. Domínguez and J. Tojo, *J. Chem. Thermodyn.*, **36**, 267 (2004); https://doi.org/10.1016/j.jct.2003.12.005.
24. M.I. Aralaguppi and J.G. Baragi, *J. Chem. Thermodyn.*, **38**, 434 (2006); https://doi.org/10.1016/j.jct.2005.06.011.
25. R.A. Clará, A.C. Gómez Marigliano and H.N. Sólimo, *J. Chem. Thermodyn.*, **40**, 292 (2008); https://doi.org/10.1016/j.jct.2007.06.009.
26. A. Toumi and M. Bouanz, *J. Mol. Liq.*, **139**, 55 (2008); https://doi.org/10.1016/j.molliq.2007.11.005.
27. M.N.M. Al-Hayan and M.A.M. Al-Bader, *J. Chem. Thermodyn.*, **38**, 1100 (2006); https://doi.org/10.1016/j.jct.2005.11.012.