Calculation of solubility of N-ethylcarbazole in ethanol+petroleum ether mixtures at various temperatures

Abolghasem Jouyban[*,**] **and William Eugence Acree Jr.**[***,†]

*Pharmaceutical Analysis Research Center and Faculty of Pharmacy, Tabriz University of Medical Sciences, Tabriz 51664, Iran
**Kimia Idea Pardaz Azarbayjan (KIPA) Science Based Company, Tabriz University of Medical Sciences, Tabriz 51664, Iran
***Department of Chemistry, University of North Texas, Denton, TX 76203-5070, U.S.A.

Abstract−Published solubility data for N-ethylcarbazole dissolved in binary (ethanol+petroleum ether) solvent mixtures has been reanalyzed by using the combined nearly ideal binary solvent/Redlich-Kister (CNIBS/R-K) and Jouyban-Acree models. The CIBS/R-K model was found to describe the observed solubility data to within an overall absolute mean deviation of 1.7%. Slightly larger mean deviations of approximately 6.6% were noted for the Jouyban-Acree model. Mathematical expressions are derived for predicting the solubility N-ethylcarbazole dissolved in binary (ethanol+petroleum ether) solvent mixtures as a function of both temperature and binary solvent composition.

Keywords: N-ethylcarbazole, Solubility Predictions, Binary Solvent Mixtures

In a recent paper appearing in this *Journal* [1], Yang and coworkers reported the experimental mole fraction solubility of N-ethylcarbazole dissolved in binary (ethanol+petroleum ether) solvent mixtures in the temperature range from 280.15 K to 316.15 K. We compared the abilities of the modified versions of the combined nearly ideal binary solvent/Redlich-Kister (CNIBS/R-K) equation:

$$\ln x_1^{sat} = x_2 \ln(x_1^{sat})_2 + x_3 \ln(x_1^{sat})_2 + x_2 x_3 \sum_{i=0}^{N} A_i (x_2 - x_3)^i \qquad (1)$$

and the Jouyban-Acree model:

$$\ln x_1^{sat} = x_2 \ln(x_1^{sat})_2 + x_3 \ln(x_1^{sat})_2 + x_2 x_3 \sum_{i=0}^{N} \frac{J_i (x_2 - x_3)^i}{T} \qquad (2)$$

to mathematically represent the measured solubility data. In Eqs. (1) and (2) (x_1^{sat}) denotes the mole fraction solubility of the solute in pure solvent i at the system temperature T, x_2 and x_3 are the initial mole fraction compositions of the solvent components 2 and 3, respectively, and A_i and J_i represent the model curve-fit parameters calculated from a regression analysis using the measured mole fraction solubility. The authors calculated the numerical values of the various curve-fit parameters by expanding Eqs. (1) and (2) into a power-series in x_2. The purpose of the present communication is to point out several shortcomings and mathematical errors in the published paper, and to provide a better mathematical representation of the measured solubility data.

First, the mean absolute deviations that are listed in the last column of Table 4 of Reference [1] are not consistent with the individual relative percent deviations that are listed in Table 2 of Reference [1] for each of the measured solubilities. For example, at T=288.15

K the authors give in Table 2 Reference [1] the values of 12.399, 9.582, 3.651, 10.256, 20.264, 28.583, 36.993, 42.777, 44.465, 47.052, and 48.520 for $100((x_1 - x_1^{cal})/x_1)$, with the calculated values being based on Eq. (12) in the authors' paper. Eq. (12) of Reference [1] is presumably derived from the Jouyban-Acree model; however, as will be shown shortly, the derivation is incorrect. The mean absolute deviation that the authors give in the last column of Table 4 of Reference [1] for T=288.15 K is 2.727, which is not close to the average of the 11 individual percent relative deviations listed above. In fact, the value of 2.727 is not even between the smallest (3.651) and largest (48.520) of the individual values. Clearly the authors' values are not consistent. There are other inconsistencies in the tabulated numerical values in Tables 2 and 4 of Reference [1] for the other temperatures studied.

Second, the authors list an overall mean absolute relative deviation of 1.957, calculated as MD=$100((\sum |x_1 - x_1^{cal}|/x_1)/N)$, near the bottom of Table 4 of Reference [1]. One would assume that the placement of 1.957 as the last numerical entry in the last column of Table 4 of Reference [1] means that the value pertains to the 10 MD values listed immediately above. The overall numerical value of 1.957 is not the mean of the 10 values listed above it. We have absolutely no idea of how the authors calculated this value.

As mentioned, we also have concerns regarding the authors' mathematical manipulation of the Jouyban-Acree expression to get the power series expansion in x_2. Their manipulation starts by replacing x_3 in the Jouyban-Acree equation with $x_3 = 1 - x_2$, and for N=2; the following expression was obtained:

$$\ln x_1 = \ln(x_1^{sat})_3 + (\ln(x_1^{sat})_2 - \ln(x_1^{sat})_3)x_2 + \frac{(J_0 - J_1 + J_2)x_2}{T}$$

$$+ \frac{(-J_0 + 3J_1 - 5J_2)x_2^2}{T} + \frac{(-J_1 + 8J_2)x_2^3}{T} + \frac{(-4J_2)x_2^4}{T} \qquad (3)$$

The authors state that Eq. (3) (numbered Eq. (11) in Reference [1])

†To whom correspondence should be addressed.
E-mail: acree@unt.edu

can be further simplified as:

$$T \ln x_1 = A_0 + A_1 T + A_2 T \ x_2 + A_3 x_2 + A_4 x_2^2 + A_5 x_2^3 + A_6 x_2^4 \quad (4)$$

where A_0, A_1, A_2, A_3, A_4, A_5 and A_6 are the model constants. There is no way to get the A_0 term simply by multiplying both sides of Eq. (3) of this paper by T.

The way to get the A_0 term into the mathematical representation would be to assume a van't Hoff type equation

$$\ln(x_1^{sat})_i = A + \frac{B}{T} \quad (5)$$

for the variation of the mole fraction solubility in each pure solvent with temperature; however, the authors made no mention in the paper that this was done. The modified Apelblat equation

$$\ln(x_1^{sat})_i = A + \frac{B}{T} + C \ln T \quad (6)$$

would provide perhaps a better mathematical description for the variation in solubility with temperature.

We have reanalyzed the experimental N-ethylcarbazole solubility data reported by Yang et al. [1] in a slightly different fashion. For the CNIBS/R-K model we reduced the number of curve-fit equation coefficients by fitting the measured solubility data to Eq. (1) of this paper using a no intercept least square analysis as explained in the literature [2]. The primary difference between our analysis and that of the authors is that we have reduced the number of curve-fit equation coefficients from 5 to 3 by retaining the $x_2 \ln(x_1^{sat})_2 + x_3 \ln(x_1^{sat})_3$ terms in the model. Our calculated model coefficients are given in Table 1 of this paper, along with the respective MD values. As expected, our MD values are slightly larger than those reported by Yang et al. [1]. Generally, the deviations between the measured solubility data and back-calculated values generally increase as one reduces the number of curve-fit parameters.

We also note that in expanding the CNIBS/R-K model to a power series expansion in x_2, Yang et al. [1] derived equation (Eq. (4) in Reference [1]):

$$\ln x_1^{sat} = B_0 + B_1 x_2 + B_2 x_2^2 + B_3 x_2^3 + B_4 x_2^4 \quad (7)$$

Table 1. Recalculated CNIBS/R-K model constants based on Eq. (1) of this paper using a no intercept least squares analysis

T	S_0	S_1	S_2	MD
280.15	2.362	1.618	0.707	0.288
284.15	2.507	1.974	1.515	1.912
288.15	2.584	2.042	1.768	0.641
292.15	2.503	1.573	1.762	0.247
296.15	2.816	2.360	1.050	0.436
300.15	3.118	2.506	2.225	1.260
304.15	2.496	2.436	5.526	4.351
308.15	2.248	2.133	4.469	3.257
312.15	1.942	1.783	3.488	2.412
316.15	1.712	1.532	2.834	1.879
			Overall MD	1.668

Table 2. Recalculated curve-fit model coefficients for Eq. (7) of this paper with the B_4-coefficient removed

T	B_0	B_1	B_2	B_3	MD
280.15	−3.503	−0.756	2.895	−3.518	0.9669
284.15	−3.247	−1.106	3.972	−4.322	2.2479
288.15	−2.989	−1.391	4.653	−4.866	2.7146
292.15	−2.758	−1.023	3.362	−3.954	2.6454
296.15	−2.555	−1.432	4.917	−5.179	1.6338
300.15	−2.347	−1.503	5.586	−5.836	3.0462
304.15	−2.134	−2.349	7.462	−6.698	7.6019
308.15	−1.900	−2.021	6.378	−5.807	6.1417
312.15	−1.828	−1.662	5.182	−4.795	4.7919
316.15	−1.764	−1.409	4.345	−4.077	3.8931
				Overall MD	3.56834

Table 3. Summarized comparison between the measured mole fraction solubility of N-ethylcarbazole in binary (ethanol+petroleum ether) mixtures and calculated values based on the CNIBS/R-K model with model curve-fit parameters determined by different regression methods

T	x_2	x_1^{sat}	x_1^{sat}-CNIBS/R-K[a]	ID-CNIBS/R-K[a]	ID-Eq. (4)- reported[b]	Recalculated ID-Eq. (4)[c]	ID-GCM calculated[d]
280.15	0.000	0.03001	0.03001	0	0.003	−0.022	−0.312
280.15	0.320	0.02881	0.02886	−0.172	−0.014	−20.908	1.658
280.15	0.515	0.02684	0.02672	0.436	0.018	−4.046	−1.302
280.15	0.645	0.02361	0.02366	−0.218	0.140	0.113	−1.621
280.15	0.739	0.02021	0.02022	−0.059	−0.114	0.403	−0.143
280.15	0.809	0.01699	0.01702	−0.182	−0.571	−0.675	0.702
280.15	0.864	0.01424	0.01425	−0.046	0.429	−1.251	1.185
280.15	0.908	0.01191	0.01197	−0.474	0.214	−1.769	0.532

[a]Calculated mole fraction solubility of N-ethylcarbazole and individual deviations (ID) based on the CNIBS/R-K model and the calculated equation coefficients given in Table 1 of this communication

[b]Individual deviations reported by Yang et al. [1] in the third column of Table 2 of Reference [1]

[c]Individual deviations that we calculate based on the equation coefficients reported by Yang et al. [1] in Table 3 of Reference [1]

[d]Individual deviations that we calculate based on the general cosolvency model and the equation coefficients reported in Table 2 of this communication

Table 3. Continued

T	x_2	x_1^{sat}	x_1^{sat}-CNIBS/R-K[a]	ID-CNIBS/R-K[a]	ID-Eq. (4)- reported[b]	Recalculated ID-Eq. (4)[c]	ID-GCM calculated[d]
280.15	0.944	0.01021	0.01013	0.807	0.284	0.073	1.124
280.15	0.974	0.00872	0.00865	0.771	1.388	1.089	0.082
280.15	1.000	0.00744	0.00744	0	−2.254	1.795	−1.976
284.15	0.000	0.03872	0.03872	0	0.005	−0.040	−0.429
284.15	0.320	0.03622	0.03681	−1.636	−0.024	−16.496	1.779
284.15	0.515	0.03505	0.03373	3.753	−0.046	−0.066	0.285
284.15	0.645	0.03023	0.03041	−0.602	0.463	0.290	−3.132
284.15	0.739	0.02575	0.02633	−2.249	0.070	−0.151	−1.965
284.15	0.809	0.02152	0.02215	−2.921	−0.831	−1.316	−0.781
284.15	0.864	0.01825	0.01830	−0.293	0.663	0.126	2.169
284.15	0.908	0.01535	0.01504	1.991	1.772	1.099	3.515
284.15	0.944	0.01275	0.01239	2.847	1.506	0.693	2.516
284.15	0.974	0.01077	0.01026	4.733	2.423	1.475	1.893
284.15	1.000	0.00853	0.00853	0	−3.778	−4.516	−6.263
288.15	0.000	0.04992	0.04992	0	0.002	−0.033	−0.803
288.15	0.320	0.04612	0.04644	−0.684	−0.027	−11.054	4.024
288.15	0.515	0.04238	0.04157	1.913	0.125	0.158	−2.484
288.15	0.645	0.03672	0.03713	−1.118	−0.163	−0.168	−4.870
288.15	0.739	0.03163	0.03193	−0.953	−0.016	0.049	−1.323
288.15	0.809	0.02657	0.02665	−0.308	0.041	0.009	1.821
288.15	0.864	0.02186	0.02181	0.218	−0.082	−0.119	3.313
288.15	0.908	0.01794	0.01773	1.165	0.513	0.457	3.811
288.15	0.944	0.01451	0.01443	0.576	−0.055	−0.206	1.710
288.15	0.974	0.01179	0.01180	−0.117	−0.373	−0.706	−1.360
288.15	1.000	0.00969	0.00969	0	−0.206	−0.166	−4.342
292.15	0.000	0.06289	0.06289	0	−0.002	−0.037	−0.853
292.15	0.320	0.05932	0.05948	−0.272	0.024	−6.769	4.463
292.15	0.515	0.05137	0.05095	0.819	−0.099	−0.079	−3.631
292.15	0.645	0.04402	0.04430	−0.626	0.052	0.038	−4.410
292.15	0.739	0.03761	0.03771	−0.277	0.170	0.219	−0.700
292.15	0.809	0.03152	0.03151	0.036	0.048	−0.002	2.124
292.15	0.864	0.02600	0.02598	0.095	−0.300	−0.346	3.203
292.15	0.908	0.02139	0.02134	0.229	−0.262	−0.345	2.962
292.15	0.944	0.01763	0.01758	0.295	0.051	−0.152	1.553
292.15	0.974	0.01456	0.01457	−0.063	0.158	−0.169	−1.117
292.15	1.000	0.01212	0.01212	0	0.412	0.409	−4.082
296.15	0.000	0.07730	0.07730	0	0.001	−0.006	−0.470
296.15	0.320	0.07023	0.07054	−0.441	−0.009	−7.177	2.365
296.15	0.515	0.06653	0.06572	1.219	0.033	0.031	−1.384
296.15	0.645	0.05775	0.05813	−0.666	−0.021	−0.082	−2.857
296.15	0.739	0.04845	0.04875	−0.624	−0.041	−0.026	−0.832
296.15	0.809	0.03963	0.03973	−0.250	0.003	−0.127	1.023
296.15	0.864	0.03199	0.03193	0.200	0.006	−0.129	2.047
296.15	0.908	0.02577	0.02564	0.493	0.050	−0.104	2.072
296.15	0.944	0.02088	0.02073	0.736	0.350	0.076	1.389
296.15	0.974	0.01689	0.01692	−0.165	−0.296	−0.717	−0.933
296.15	1.000	0.01390	0.01390	0	−0.223	−0.287	−2.598
300.15	0.000	0.09480	0.09480	0	0.003	0.001	−0.907
300.15	0.320	0.09063	0.09155	−1.019	−0.035	−8.878	4.531
300.15	0.515	0.08493	0.08283	2.468	0.174	0.223	−2.941
300.15	0.645	0.07355	0.07418	−0.854	−0.326	−0.331	−5.217

Table 3. Continued

T	x_2	x_1^{sat}	x_1^{sat}-CNIBS/R-K[a]	ID-CNIBS/R-K[a]	ID-Eq. (4)- reported[b]	Recalculated ID-Eq. (4)[c]	ID-GCM calculated[d]
300.15	0.739	0.06240	0.06302	−0.999	0.245	0.322	−1.136
300.15	0.809	0.05087	0.05135	−0.938	0.187	0.136	1.891
300.15	0.864	0.04018	0.04069	−1.260	−0.836	−0.904	2.562
300.15	0.908	0.03195	0.03188	0.225	−0.182	−0.320	3.173
300.15	0.944	0.02586	0.02495	3.519	2.313	2.081	4.210
300.15	0.974	0.02015	0.01963	2.580	0.616	0.228	0.031
300.15	1.000	0.01549	0.01549	0	−3.253	−3.310	−6.910
304.15	0.000	0.11599	0.11599	0	0.003	−0.025	−2.046
304.15	0.320	0.10641	0.11018	−3.539	−0.031	−15.993	9.560
304.15	0.515	0.09683	0.08966	7.409	0.115	−11.783	−5.703
304.15	0.645	0.08725	0.08696	0.329	−0.090	−7.277	−10.202
304.15	0.739	0.07767	0.08235	−6.029	−0.146	−0.447	−5.925
304.15	0.809	0.06809	0.07249	−6.469	0.021	7.061	0.981
304.15	0.864	0.05851	0.05967	−1.975	0.200	13.810	7.169
304.15	0.908	0.04893	0.04690	4.154	0.359	18.260	10.53
304.15	0.944	0.03935	0.03586	8.858	−0.348	19.160	9.592
304.15	0.974	0.02977	0.02706	9.099	0.905	14.400	1.724
304.15	1.000	0.02019	0.02019	0	−1.575	−1.569	−20.184
308.15	0.000	0.14714	0.14714	0	−0.001	6.819	−1.701
308.15	0.320	0.13543	0.13910	−2.708	0.007	−11.249	8.073
308.15	0.515	0.12371	0.11653	5.800	−0.025	−24.575	−4.901
308.15	0.645	0.11200	0.11186	0.127	0.063	2.169	−8.476
308.15	0.739	0.10028	0.10499	−4.692	−0.020	21.430	−4.743
308.15	0.809	0.08857	0.09286	−4.840	−0.062	27.380	1.078
308.15	0.864	0.07686	0.07783	−1.258	0.020	22.600	6.196
308.15	0.908	0.06514	0.06293	3.400	0.008	6.604	8.786
308.15	0.944	0.05343	0.04983	6.732	−0.010	−25.159	7.607
308.15	0.974	0.04171	0.03909	6.274	−0.016	−85.091	0.576
308.15	1.000	0.03042	0.03042	0	0.014	−203.940	−15.421
312.15	0.000	0.15853	0.15853	0	0.001	0.019	−1.348
312.15	0.320	0.14668	0.14965	−2.024	−0.009	−5.556	6.492
312.15	0.515	0.13482	0.12888	4.410	0.022	−33.716	−3.985
312.15	0.645	0.12297	0.12293	0.030	−0.031	−37.160	−6.690
312.15	0.739	0.11112	0.11503	−3.520	0.036	−24.824	−3.613
312.15	0.809	0.09927	0.10280	−3.551	−0.092	−9.144	1.017
312.15	0.864	0.08741	0.08815	−0.850	0.083	4.718	5.010
312.15	0.908	0.07556	0.07360	2.600	0.123	14.830	6.928
312.15	0.944	0.06371	0.06053	4.986	−0.042	20.860	5.792
312.15	0.974	0.05185	0.04948	4.563	−0.123	22.500	0.231
312.15	1.000	0.04023	0.04023	0	−0.049	19.420	−11.605
316.15	0.000	0.16945	0.16945	0	−0.001	−8.242	−1.107
316.15	0.320	0.15751	0.16001	−1.588	0.004	−90.256	5.392
316.15	0.515	0.14556	0.14047	3.500	−0.018	−95.912	−3.343
316.15	0.645	0.13362	0.13364	−0.015	0.035	−90.104	−5.480
316.15	0.739	0.12167	0.12504	−2.773	−0.021	−70.409	−2.882
316.15	0.809	0.10973	0.11274	−2.747	−0.026	−44.056	0.937
316.15	0.864	0.09778	0.09837	−0.605	0.014	−17.675	4.182
316.15	0.908	0.08584	0.08406	2.074	0.065	4.502	5.663
316.15	0.944	0.07389	0.07103	3.864	−0.071	21.410	4.597
316.15	0.974	0.06195	0.05978	3.499	0.015	33.310	0.059
316.15	1.000	0.05012	0.05012	0	0.037	41.240	−9.183

A. Jouyban and W. E. Acree Jr.

Namely, the general cosolvency model (GCM) without referring to the original reference [3] and reported significant and non-significant curve-fitting parameters. The B_4 constants are not statistically significant (p>0.10). The recalculated curve-fit equation coefficients are given in Table 2 of this paper with the B_4-coefficient removed. During our reanalysis of the N-ethylcarbazole solubility data we found that there were several miscalculated individual deviations (ID) in Table 2 of Reference [1] pertaining to the modified version of the CNIBS/R-K model. We recalculated the ID values based on tabulated curve-fit equation coefficients in Table 3 of Reference [1], along with the ID values based on the curve-fit equation

coefficients from Tables 1 and 2. Our computations show that several of the IDs reported by Yang et al. pertaining to the CNIBS/R-K model were miscalculated assuming that the equation coefficients given by the authors are correct.

Our reanalysis of the N-ethylcarbazole solubility data also showed that there are many inconsistencies in the calculations pertaining to the Jouyban-Acree model. As noted above, the individual deviations given in the last column of Table 2 of reference [1] were not consistent with the MD values that the authors reported in the last column of Table 4 for each of the ten temperatures studied. Because of the noted inconsistencies we have recalculated all of individual

Table 4. Summarized comparison between the measured mole fraction solubility of N-ethylcarbazole in binary (ethanol+petroleum ether) mixtures and calculated values based on the Jouyban-Acree model with model curve-fit parameters determined by different regression methods

T	x_2	x_1^{sat}	x_1^{sat}-Jouyban-Acree model[a]	ID-Jouyban-Acree model[a]	Reported ID values[b]	Recalculated ID values[c]
280.15	0.000	0.03001	0.03001	0	10.639	11.76
280.15	0.320	0.02881	0.03077	−6.803	23.348	8.33
280.15	0.515	0.02684	0.02837	−5.702	18.193	15.13
280.15	0.645	0.02361	0.02672	−13.164	26.556	25.53
280.15	0.739	0.02021	0.02408	−19.128	33.653	36.07
280.15	0.809	0.01699	0.02072	−21.938	43.396	44.41
280.15	0.864	0.01424	0.01722	−20.937	50.873	51.11
280.15	0.908	0.01191	0.01405	−17.966	57.878	56.08
280.15	0.944	0.01021	0.01137	−11.398	63.003	60.66
280.15	0.974	0.00872	0.00920	−5.551	66.663	63.85
280.15	1.000	0.00744	0.00744	0	66.623	66.24
284.15	0.000	0.03872	0.03872	0	11.253	12.22
284.15	0.320	0.03622	0.03798	−4.846	17.725	3.56
284.15	0.515	0.03505	0.03412	2.640	11.542	−2.28
284.15	0.645	0.03023	0.03159	−4.511	15.455	20.25
284.15	0.739	0.02575	0.02814	−9.298	19.321	30.29
284.15	0.809	0.02152	0.02404	−11.694	36.469	38.35
284.15	0.864	0.01825	0.01988	−8.940	45.503	45.91
284.15	0.908	0.01535	0.01617	−5.330	51.743	51.29
284.15	0.944	0.01275	0.01306	−2.453	56.054	54.65
284.15	0.974	0.01077	0.01056	1.962	59.939	57.60
284.15	1.000	0.00853	0.00853	0	59.336	57.10
288.15	0.000	0.04992	0.04992	0	12.399	13.22
288.15	0.320	0.04612	0.04671	−1.288	9.582	0.61
288.15	0.515	0.04238	0.04085	3.616	3.651	2.90
288.15	0.645	0.03672	0.03714	−1.141	10.256	10.76
288.15	0.739	0.03163	0.03268	−3.323	20.264	21.88
288.15	0.809	0.02657	0.02768	−4.196	28.583	30.53
288.15	0.864	0.02186	0.02277	−4.181	36.993	36.59
288.15	0.908	0.01794	0.01845	−2.862	42.777	41.01
288.15	0.944	0.01451	0.01487	−2.509	44.465	43.20
288.15	0.974	0.01179	0.01201	−1.824	47.052	44.46
288.15	1.000	0.00969	0.00969	0	48.520	45.56

[a]Calculated mole fraction solubility of N-ethylcarbazole and individual deviations (ID) based on the Jouyban-Acree model and the calculated coefficients given in Eq. (8) of this communication
[b]ID values that were reported in Table 2 of Reference [1]
[c]Recalculated ID values based on the model constants given in Table 4 of Reference [1]

Table 4. Continued

T	x_2	x_1^{sat}	x_1^{sat}-Jouyban-Acree model[a]	ID-Jouyban-Acree model[a]	Reported ID values[b]	Recalculated ID values[c]
292.15	0.000	0.06289	0.06289	0	12.105	12.77
292.15	0.320	0.05932	0.05835	1.638	4.885	−0.65
292.15	0.515	0.05137	0.05082	1.070	−6.179	−6.37
292.15	0.645	0.04402	0.04609	−4.698	−0.609	−0.33
292.15	0.739	0.03761	0.04051	−7.721	9.496	10.34
292.15	0.809	0.03152	0.03433	−8.906	16.759	19.25
292.15	0.864	0.02600	0.02827	−8.721	25.091	25.83
292.15	0.908	0.02139	0.02294	−7.250	29.827	30.64
292.15	0.944	0.01763	0.01853	−5.082	37.698	34.02
292.15	0.974	0.01456	0.01498	−2.906	43.678	36.15
292.15	1.000	0.01212	0.01212	0	37.772	37.88
296.15	0.000	0.07730	0.07730	0	10.17	10.71
296.15	0.320	0.07023	0.06972	0.728	−3.307	−9.95
296.15	0.515	0.06653	0.05976	10.170	−8.573	−8.23
296.15	0.645	0.05775	0.05363	7.141	−2.091	−2.25
296.15	0.739	0.04845	0.04682	3.369	5.279	5.82
296.15	0.809	0.03963	0.03950	0.320	13.587	12.21
296.15	0.864	0.03199	0.03245	−1.443	1.902	16.88
296.15	0.908	0.02577	0.02630	−2.075	25.975	20.02
296.15	0.944	0.02088	0.02123	−1.695	29.074	22.11
296.15	0.974	0.01689	0.01718	−1.689	28.443	22.60
296.15	1.000	0.01390	0.01390	0	24.145	23.42
300.15	0.000	0.09480	0.09480	0	8.554	8.96
300.15	0.320	0.09063	0.08243	9.047	−1.270	−9.44
300.15	0.515	0.08493	0.06918	18.540	−10.222	−10.9
300.15	0.645	0.07355	0.06122	16.760	−10.097	−6.51
300.15	0.739	0.0624	0.05295	15.140	2.626	1.84
300.15	0.809	0.05087	0.04441	12.700	5.687	7.29
300.15	0.864	0.04018	0.03634	9.559	8.689	9.53
300.15	0.908	0.03195	0.02938	8.029	11.702	11.16
300.15	0.944	0.02586	0.02369	8.405	15.114	12.84
300.15	0.974	0.02015	0.01915	4.987	18.070	9.58
300.15	1.000	0.01549	0.01549	0	7.036	3.75
304.15	0.000	0.11599	0.11599	0	7.223	7.50
304.15	0.320	0.10641	0.10232	3.839	−3.251	−18.95
304.15	0.515	0.09683	0.08674	10.420	−12.153	−26.34
304.15	0.645	0.08725	0.07728	11.430	−10.954	−18.24
304.15	0.739	0.07767	0.06721	13.460	−6.501	−5.05
304.15	0.809	0.06809	0.05666	16.790	−2.014	6.85
304.15	0.864	0.05851	0.04659	20.380	1.450	15.76
304.15	0.908	0.04893	0.03784	22.660	3.841	20.78
304.15	0.944	0.03935	0.03063	22.150	4.549	21.29
304.15	0.974	0.02977	0.02486	16.500	6.458	15.45
304.15	1.000	0.02019	0.02019	0	4.353	−2.51
308.15	0.000	0.14714	0.14714	0	2.627	9.86
308.15	0.320	0.13543	0.13638	−0.701	−2.099	−18.50
308.15	0.515	0.12371	0.11928	3.578	−15.163	−27.57
308.15	0.645	0.11200	0.10851	3.114	14.940	−20.43
308.15	0.739	0.10028	0.09589	4.377	−11.659	−7.58
308.15	0.809	0.08857	0.08186	7.574	−8.678	4.44
308.15	0.864	0.07686	0.06804	11.480	−5.543	13.73

Table 4. Continued

T	x_2	x_1^{sat}	x_1^{sat}-Jouyban-Acree model[a]	ID-Jouyban-Acree model[a]	Reported ID values[b]	Recalculated ID values[c]
308.15	0.908	0.06514	0.05578	14.370	−2.104	19.40
308.15	0.944	0.05343	0.04552	14.800	−3.832	21.01
308.15	0.974	0.04171	0.03721	10.790	−4.959	17.33
308.15	1.000	0.03042	0.03042	0	80.477	6.34
312.15	0.000	0.15853	0.15853	0	−2.799	−2.86
312.15	0.320	0.14668	0.15604	−6.380	−32.500	−37.90
312.15	0.515	0.13482	0.14173	−5.123	−8.715	−50.03
312.15	0.645	0.12297	0.13222	−7.524	−8.505	−42.43
312.15	0.739	0.11112	0.11908	−7.163	−0.908	−27.44
312.15	0.809	0.09927	0.10319	−3.949	−8.768	−12.95
312.15	0.864	0.08741	0.08685	0.646	−4.987	−1.27
312.15	0.908	0.07556	0.07197	4.758	−5.087	6.60
312.15	0.944	0.06371	0.05928	6.956	−5.790	10.44
312.15	0.974	0.05185	0.04885	5.793	−12.715	9.63
312.15	1.000	0.04023	0.04023	0	−32.868	3.31
316.15	0.000	0.16945	0.16945	0	−8.539	−17.69
316.15	0.320	0.15751	0.17423	−10.616	15.267	−60.90
316.15	0.515	0.14556	0.16270	−11.773	9.736	−76.98
316.15	0.645	0.13362	0.15463	−15.722	11.249	−69.09
316.15	0.739	0.12167	0.14123	−16.079	9.435	−51.74
316.15	0.809	0.10973	0.12378	−12.805	5.114	−34.39
316.15	0.864	0.09778	0.10519	−7.574	−3.600	−19.98
316.15	0.908	0.08584	0.08789	−2.393	−7.573	−9.67
316.15	0.944	0.07389	0.07293	1.296	−90.927	−3.61
316.15	0.974	0.06195	0.06049	2.354	−19.182	−1.99
316.15	1.000	0.05012	0.05012	0	−39.211	−5.13

deviations using the calculated curve-fit equation coefficients determined by Yang et al. [1]. We have also analyzed the solubility data in accordance with the original Jouyban-Acree model, and obtained the following expression:

$$\ln x_{1,T}^{sat} = x_2 \ln(x_1^{sat})_{2,T} + x_3 \ln(x_1^{sat})_{3,T} \qquad (8)$$
$$+ x_2 x_3 \left(\frac{724.122}{T} + \frac{592.9345(x_2 - x_3)}{T} + \frac{741.5465(x_2 - x_3)^2}{T} \right)$$

where $(x_1^{sat})_{i,T}$ denotes the mole fraction solubility of the solute in pure solvent i at the system temperature T. The results of our computations are given in Tables 4 and 5. Examination of the numerical entries reveals that that the descriptive ability of the Jouyban-Acree model (when properly applied) is far better than what one would be led to believe based on the individual deviations reported

by Yang et al. [1]. Examination further reveals that we were unable to validate the calculations of Yang et al. using the equation coefficients given by the authors in Table 4 of their paper. We recommend the researchers in the field to use Eq. (8) of this paper instead of Eq. (7) of this paper, since it possesses more rigorous theoretical justifications on derivation [4,5] and could be used to extend the prediction capability to other solutes and solvents systems employing some physico-chemical properties of the solutes and solvents in the computations. For examples on such applications, readers could refer to generally trained models presented in earlier works [6,7].

One final note: A more relevant analysis of the solubility of a solute in binary solvent mixtures at various temperatures is the combined version of the Jouyban-Acree and van't Hoff models [8]:

Table 5. Descriptive statistics for the Jouyban-Acree Model with different numerical values of the calculated curve-fit equation coefficients

MD	N	Minimum	Maximum	Mean	Std. deviation
Classical Jouyban-Acree model	110	.00	22.66	6.5119	6.17201
Mean of reported IDs in Table 2 of Reference [1]	110	.61	90.93	19.8520	19.51711
Published MD in Table 4 of Reference [1]	110	1.96	1.96	1.9570	
MD of re-calculated data using constants reported in Table 4 of Reference [1]	110	.33	76.98	22.3144	18.49734
MD of re-fitted data to Eq. (12) of Reference [1]	110	.09	34.09	7.2487	6.07836

$$\ln x_{1,T}^{sat} = x_2\left(K_1 + \frac{K_2}{T}\right) + x_3\left(K_3 + \frac{K_4}{T}\right) + x_2 x_3 \sum_{i=0}^{N} \frac{J_i(x_2 - x_3)^i}{T} \qquad (9)$$

The trained version is:

$$\ln x_{1,T}^{sat} = x_2\left(13.035 - \frac{5084.878}{T}\right) + x_3\left(11.996 - \frac{4319.812}{T}\right)$$
$$+ 715.098\left(\frac{x_2 x_3}{T}\right) + 576.371\frac{x_2 x_3 (x_2 - x_3)}{T} \qquad (10)$$
$$+ 641.445\frac{x_2 x_3 (x_2 - x_3)^2}{T}$$

which reproduces the experimental data with MD of 6.76%. The main advantage of Eq. (10) in this paper over Eq. (8) is that it does not require any more experimental data for predicting the solubility of N-ethylcarbazole in any solvent composition of ethanol+petroleum ether at different T, whereas Eq. (8) requires the experimental solubility data in the mono-solvents at each temperature of interest, i.e. $(x_1^{sat})_{2,T}$ and $(x_1^{sat})_{3,T}$ values. We believe that more accurate experimental data and accurate computations using more reliable and predictive models are required in the industry for facilitating process design procedures.

REFERENCES

1. W. Yang, K. Wu, Y. Hu, T. Zhang, Q. Guo, S. Yang and Y. Shi, *Korean J. Chem. Eng.*, **32**, 1158 (2015).
2. A. Jouyban-Gharamaleki and J. Hanaee, *Int. J. Pharm.*, **154**, 245 (1997).
3. M. Barzegar-Jalali and A. Jouyban-Gharamaleki, *Int. J. Pharm.*, **152**, 247 (1997).
4. W. E. Acree Jr., *Thermochim. Acta*, **198**, 71 (1992).
5. A. Jouyban-Gharamaleki and W. E. Acree Jr., *Int. J. Pharm.*, **167**, 177 (1998).
6. A. Jouyban, A. Shayanfar and W. E. Acree Jr., *Fluid Phase Equilib.*, **293**, 47 (2010).
7. A. Jouyban, Sh. Soltanpour, S. Soltani, E. Tamizi, M. A. A. Fakhree and W. E. Acree Jr., *J. Mol. Liq.*, **146**, 82 (2009).
8. A. Jouyban, M. A. A. Fakhree and W. E. Acree Jr., *J. Chem. Eng. Data*, **57**, 1344 (2012).