Error Analysis of Linearization Methods in Regression of Data for the Van Laar and Margules Equations

Mordechai Shacham* and Jaime Wisniak

Department of Chemical Engineering, Ben-Gurion University of the Negev, Beer-Sheva, 84105 Israel

Neima Brauner

School of Engineering, Tel-Aviv University, Tel-Aviv, 69978 Israel

Linearization is used extensively in regression and model testing of experimental data. In this work, Margules and Van Laar equation parameters have been calculated, for one sample data set, using several different linearization methods. The resultant parameter values were compared with values obtained by nonlinear regression. The increase in absolute error and change of its distribution due to the transformation of the data were also analyzed. It is concluded that linearization may lead to inaccurate or even completely incorrect parameter values which do not describe the original data adequately. The commonly used statistical tests may not detect the inaccuracy of the calculated parameter values. The ultimate test is the accurate recovery of the original activity coefficient data.

Introduction

Linearization is used extensively in regression and model testing of experimental data, because it can be carried out graphically and also because linear regression software is more readily available than nonlinear software. Unfortunately, linearization may lead to false conclusions (Wisniak, 1993), and the statistical tests used to check the goodness of fit will often not detect that the parameters are incorrect.

We have used the Margules and Van Laar equations to illustrate the problems that arise when linearization is used. The Van Laar and Margules equations are extensively used for the correlation of activity coefficients, particularly in binary solutions. They are mathematically simple and require only two adjustable parameters.

There are many ways in which these parameters can be calculated from experimental data (King, 1969). Of these, we will only analyze the ones that involve linearization of the basic equations. The accuracy of the different methods will be compared using equilibrium data for 1,1,1-trichloroethane (1) and propanol (2) at 96.7 kPa determined by Kumar and Rao (1991) and reported in Table I.

Margules Equation (Walas, 1985)

The Margules equation describes the excess Gibbs energy as follows:

$$g = G_E/RT = x_1 \ln \gamma_1 + x_2 \ln \gamma_2 = x_1x_2(Ax_2 + Bx_1) \quad (1)$$

where x_1 and x_2 are mole fractions of components 1 and 2, respectively; γ_1 and γ_2 are the activity coefficients; and A and B are two constants, independent of the temperature and composition, characteristic of the components.

The pertinent expressions for the activity coefficients are

$$\gamma_1 = \exp[x_2^2(2B - A) + 2x_2^3(A - B)] \quad (2)$$

$$\gamma_2 = \exp[x_1^2(2A - B) + 2x_1^3(B - A)] \quad (3)$$

The coefficients A and B can be determined from either eq 1 (full population of data) or eqs 2 and 3 (50% of data population), as follows:

Table I. Activity Coefficient Data

	x_1	x_2	γ_1	γ_2
1	0.0246	0.9754	5.2313	1.0036
2	0.0496	0.9504	4.9516	1.0084
3	0.0648	0.9352	5.0295	1.0153
4	0.0996	0.9004	4.6721	1.0283
5	0.1498	0.8502	3.959	1.0502
6	0.2999	0.7001	2.4264	1.1865
7	0.3718	0.6282	2.0725	1.2918
8	0.4548	0.5452	1.781	1.4699
9	0.5549	0.4451	1.549	1.7619
10	0.6397	0.3603	1.3695	2.1246
11	0.8031	0.1969	1.1698	3.2094
12	0.8698	0.1302	1.0745	4.6223
13	0.9478	0.0522	0.9998	6.5437

1. Nonlinear Regression, Full Population. The best values of A and B in the least-squares-error sense will be found by minimizing directly the nonlinear objective function:

$$\min_{A,B} S^2 = \sum_{i=1}^{n}(\gamma_{1,i}^{\mathrm{calc}} - \gamma_{1,i}^{\mathrm{obs}})^2 + \sum_{i=1}^{n}(\gamma_{2,i}^{\mathrm{calc}} - \gamma_{2,i}^{\mathrm{obs}})^2 \quad (4)$$

where n is the number of data points and $\gamma_{1,i}^{\mathrm{calc}}$ and $\gamma_{2,i}^{\mathrm{calc}}$ are calculated from eqs 2 and 3, respectively.

2. Nonlinear Regression, Half Population. Utilization of eq 4 requires the use of an objective function which is the sum of two different objective functions. Some nonlinear regression programs will not allow the use of such combined expression. Equation 4 can be separated into two different objective functions:

$$\min_{A,B} S_1^2 = \sum_{i=1}^{n}(\gamma_{1,i}^{\mathrm{calc}} - \gamma_{1,i}^{\mathrm{obs}})^2 \quad (5)$$

$$\min_{A,B} S_2^2 = \sum_{i=1}^{n}(\gamma_{2,i}^{\mathrm{calc}} - \gamma_{2,i}^{\mathrm{obs}})^2 \quad (6)$$

If there is no experimental error, it can be expected from theory that the values of A and B calculated from eqs 5 or 6 are equal. They are expected to be equal since $\gamma_{1,i}$ and $\gamma_{2,i}$ are not measured independently but calculated from the same measurements.

* Author to whom correspondence should be addressed.

0888-5885/93/2632-2820$04.00/0

3. Multiple Linear Regression, Half Population. Taking ln of both sides of eqs 2 and 3 gives

$$\ln \gamma_1 = x_2^2(2B - A) + 2x_2^3(A - B) \quad (7)$$

$$\ln \gamma_2 = x_1^2(2A - B) + 2x_1^3(B - A) \quad (8)$$

These expressions can be linearized by defining new transformation variables: $y_1 \equiv \ln \gamma_1$; $y_2 = \ln \gamma_2$; $x_1' = x_1^2$; $x_1'' = 2x_1^3$; $x_2' = x_2^2$; and $x_2'' = 2x_2^3$. Introducing the new variables into eqs 7 and 8 leads to

$$y_1 = x_2'(2B - A) + x_2''(A - B) \quad (9)$$

$$y_2 = x_1'(2A - B) + x_1''(B - A) \quad (10)$$

4. Straight Line Regression, Half Population. While eqs 9 and 10 are linear in A and B, graphical methods cannot be used to find the parameters. To allow the use of graphical methods, eq 7 can be divided by x_2^2 and eq 8 by x_1^2. Then the following transformed variables can be defined:

$$z_1 \equiv \ln \gamma_1/x_2^2; \quad z_2 \equiv \ln \gamma_2/x_1^2; \quad x_1' \equiv 2x_1; \quad x_2' \equiv 2x_2$$

Using these transformations, the following expressions are obtained:

$$z_1 = (2B - A) + x_2'(A - B) \quad (11)$$

$$z_2 = (2A - B) + x_1'(B - A) \quad (12)$$

5. Multiple Linear Regression, Full Population. The combined eq 3 is used in this case. Defining the transformed variables

$$v \equiv x_1 \ln \gamma_1 + x_2 \ln \gamma_2; \quad x_1' \equiv x_1x_2^2; \quad x_2' \equiv x_2x_1^2$$

and substituting into (1) gives

$$v = Ax_1' + Bx_2' \quad (13)$$

6. Straight Line Regression, Full Population. In this case, the transformation function

$$w \equiv \frac{x_1 \ln \gamma_1 + x_2 \ln \gamma_2}{x_1x_2}$$

is defined. Dividing both sides of eq 3 by x_1x_2, noting tht $x_1 = 1 - x_2$, leads to the following expression:

$$w = (A - B)x_2 + B \quad (14)$$

Van Laar Equation (Walas, 1985)

The pertinent equations for the Van Laar model are as follows:

$$g = G_E/RT = x_1 \ln \gamma_1 + x_2 \ln \gamma_2 = ABx_1x_2/(Ax_1 + Bx_2) \quad (15)$$

$$\gamma_1 = \exp(A/[1 + (x_1/x_2)(A/B)]^2) \quad (16)$$

$$\gamma_2 = \exp(B/[1 + (x_2/x_1)(B/A)]^2) \quad (17)$$

The coefficients A and B can be determined as follows:

1. Nonlinear Regression, Full Population. Equation 4 is used again, but γ_1^{calc} and γ_2^{calc} are calculated from eqs 16 and 17, respectively.

2. Nonlinear Regression, Half Population. Equations 5 and 6 are used again with the appropriate γ_1^{calc} and γ_2^{calc} values.

3. Multiple Linear Regression, Half Population. Multiple linear regression can be used only in an iterative fashion. There is no advantage in using it for half population; hence, this case will be discussed in connection with full population multiple regression.

4. Straight Line Regression, Half Population. By manipulating eqs 16 and 17, the following expressions can be obtained:

$$\frac{1}{(\ln \gamma_1)^{1/2}} = \frac{1}{A^{1/2}} + \frac{x_1}{x_2}\frac{A^{1/2}}{B} \quad (18)$$

$$\frac{1}{(\ln \gamma_2)^{1/2}} = \frac{1}{B^{1/2}} + \frac{x_2}{x_1}\frac{B^{1/2}}{A} \quad (19)$$

Specifying the transformed variables

$$z_1 \equiv \frac{1}{(\ln \gamma_1)^{1/2}}; \quad z_2 \equiv \frac{1}{(\ln \gamma_2)^{1/2}}; \quad x_1' = x_1/x_2; \quad x_2' = x_2/x_1$$

the following linear functions are obtained:

$$z_1 = A' + B'x_1' \quad (20)$$

$$z_2 = A'' + B''x_2' \quad (21)$$

where $A' = 1/A^{1/2}$, $B' = A^{1/2}/B$, $A'' = 1/B^{1/2}$, and $B'' = B^{1/2}/A$.

5. Multiple Linear Regression, Full Population. Equation 15 can be rewritten

$$x_1 \ln \gamma_1 + x_2 \ln \gamma_2 = x_1A/[1 + (x_1/x_2)(A/B)]^2 + x_2B/[1 + (x_2/x_1)(B/A)]^2 \quad (22)$$

Using the definition of the transformed variable $v \equiv x_1 \ln \gamma_1 + x_2 \ln \gamma_2$ the following iterative procedure can be used to find A and B: (i) estimate an initial value for the ratio $r = A/B$ ($r = 1$ can be a good estimate), (ii) define transformed variables $x_1' \equiv 1/[1 + (x_1/x_2)(r)]^2$ and $x_2' \equiv 1/[1 + (x_2/x_1)(1/r)]^2$, (iii) solve the multiple linear regression problem

$$V = Ax_1' + Bx_2' \quad (23)$$

for A and B, and (iv) calculate a new value for $r = A/B$. If it is close enough to the previous value, end the iteration, otherwise, go back to step ii.

6. Straight Line Regression, Full Population. Equation 15 can be rearranged to give

$$\frac{x_1}{x_1 \ln \gamma_1 + x_2 \ln \gamma_2} = \frac{1}{A} + \frac{1}{B}\frac{x_1}{x_2} \quad (24)$$

Defining

$$w \equiv \frac{x_1}{x_1 \ln \gamma_1 + x_2 \ln \gamma_2} \quad \text{and} \quad x_1' \equiv x_1/x_2$$

yields

$$w = A' + B'x_1' \quad (25)$$

where $A' = 1/A$ and $B' = 1/B$.

Table II. Errors in the Dependent Variables for the Margules Equation[a]

used in eq	variable	|calcd − obsd value|	relative error, %
2	γ_1	0.0335	3.12
3	γ_2	0.0767	0.4
9	$\ln \gamma_1$	0.03171	44.0
10	$\ln \gamma_2$	0.0167	1.096
11	z_1	1.877	44.0
12	z_2	0.022	1.1
13	v	0.0298	11.4
14	w	0.26	11.38

[a] $x_1 = 0.8698$, $\gamma_{1,obs} = 1.0745$, $x_2 = 0.1302$, $\gamma_{2,obs} = 4.6223$, $\gamma_{1,calc} = 1.041$, $\gamma_{2,calc} = 4.5456$ (the latter two values calculated using $A = 1.7614$ and $B = 2.11$).

Table III. Errors in the Dependent Variables for the Van Laar Equation[a]

used in eq	variable	|calcd − obsd value|	relative error, %
15	γ_1	0.0351	3.26
16	γ_2	0.1963	4.24
17	$\ln \gamma_1$	0.0332	46.0
17	$\ln \gamma_2$	0.0434	2.83
20	z_1	1.356	36.0
21	z_2	0.011	14.0
23	v	0.03458	13.2
25	w	0.505	15.2

[a] $x_1 = 0.8698$, $\gamma_{1,obs} = 1.0745$, $x_2 = 0.1302$, $\gamma_{2,obs} = 4.6223$, $\gamma_{1,calc} = 1.0394$, $\gamma_{2,calc} = 4.42592$ (the latter two values calculated using $A = 1.7667$ and $B = 2.1291$).

Error Analysis of the Transformation Functions

For the least-squares error regression to give the best parameter values, we have to satisfy the assumptions that only the dependent variables (in this case γ_1 and γ_2) are subject to experimental error and that the independent variables (x_1 and x_2 in this case) are accurate. These are reasonable assumptions in this particular case, since the liquid composition can be accurately analyzed.

The arithmetic operations used in the different linearization schemes may change the error at a particular point considerably and change the distribution of error throughout the range of the measurements. The changes in error distribution will be demonstrated first theoretically and then numerically for a particular measurement point. Let $\delta\gamma$ indicate the absolute error in the measured value of γ and $R(\gamma)$ the relative error. Thus, $R(\gamma) = |\delta\gamma|/\gamma$.

According to the general error propagation formula

$$\delta[f(x)] \leq \left|\frac{df}{dx}\right| |\delta x| \tag{26}$$

thus, the error in $\ln \gamma$ is

$$\delta(\ln \gamma) \leq \left|\frac{1}{\gamma}\right| |\delta\gamma| \tag{27}$$

and the relative error

$$R(\ln \gamma) = \frac{|\delta\gamma|}{|\gamma|\,|\ln \gamma|} \tag{28}$$

Let us investigate the case where x_1 is very close to 1 (say $x_1 > 0.9$), so γ_1 is very close to 1, say $\gamma_1 = 1 + \epsilon$ where $\epsilon < 0.1$. In this case, $\ln \gamma_1 \sim \epsilon$; thus, the relative error in $\ln \gamma_1$ is at least 10 times larger than the relative error in γ_1 itself.

If we calculate, for example, $z_1 = \ln \gamma_1/x_2^2$, the relative error will not change very much, but the absolute error will be multiplied by $1/x_2^2$ were $x_2 = 1 - x_1 < 0.1$. Thus, the error will multiply by more than 100 times.

Similar analysis can be carried out for the other transformed dependent variables (v, w, etc.). The general conclusion is that transformation of variables increases the error in them, and the change will depend on the values of x_1 and x_2. As a consequence, the absolute error distribution throughout the range of measurements ($0 < x_1 < 1$) will change.

To demonstrate the change of error introduced by the different transformations, the error has been calculated for point no. 12, where $x_1 = 0.8698$, close to 1. To estimate the exact values of γ_1 and γ_2 at these points, A and B were calculated using nonlinear regression with eq 5. These A and B values were then used to calculate $\gamma_{1,12}^{calc}$ and $\gamma_{2,12}^{calc}$ values.

Afterward, these values were used to calculate the error for various linearization methods. The results for the Margules method are summarized in Table II.

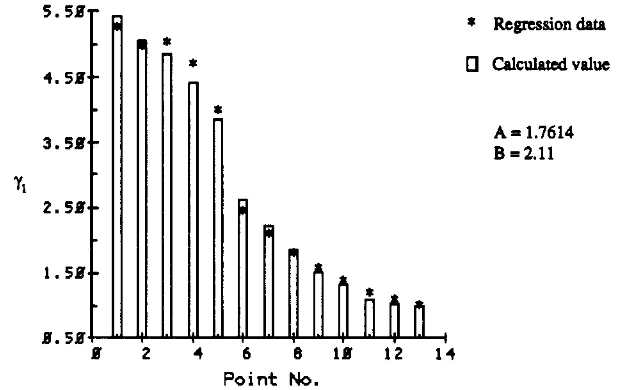

Figure 1. $\gamma_{1,obs}$ and $\gamma_{1,calc}$ when nonlinear regression with eq 5 is used.

The absolute error in γ_1 is 0.033 57, and the relative error is 3.12%. After taking the ln of γ_1, the absolute error remains about the same (0.034 71), but the relative error grows considerably, to 44%. After dividing $\ln \gamma_1$ by x_2^2 to obtain z_1, the absolute error is multiplied by more than 50 to become 1.877, while the relative error still remains 44%.

In calculating w, the error growth is more moderate because all four variables, x_1, x_2, γ_1, and γ_2, are involved. The absolute error in this case is 0.26, and the relative error 11.38%.

The results for the Van Laar equation are summarized in Table III. These results are very similar to the results obtained with the Margules equation. The absolute error increases significantly with the use of some of the transformations (z_1, for example).

Change of the Error Distribution Because of Linearization

The methods described previously were used to calculate A and B for the Margules equation.

Figures 1–4 show plots of the calculated and observed values of the variables when using the different approaches for regression of the data. The calculated constants (A and B) are also indicated.

Figures 1 and 2 show that when using nonlinear regression and plotting values of γ_1 and γ_2 the fit between the observed and calculated data is fairly good and that the differences between the calculated A and B values are not very substantial. Similar pictures are obtained when plotting $\ln \gamma_1$ and $\ln \gamma_2$. This can be surprising, considering that we have shown the relative error in $\ln \gamma_1$ can be as high as 44%. But the least-squares method minimizes the sum of squares of the absolute error and is not affected by the relative error. In addition, in graphic representation

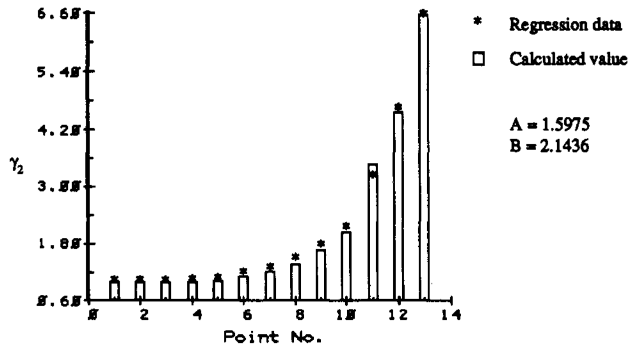

Figure 2. $\gamma_{2,obs}$ and $\gamma_{2,calc}$ when nonlinear regression with eq 6 is used.

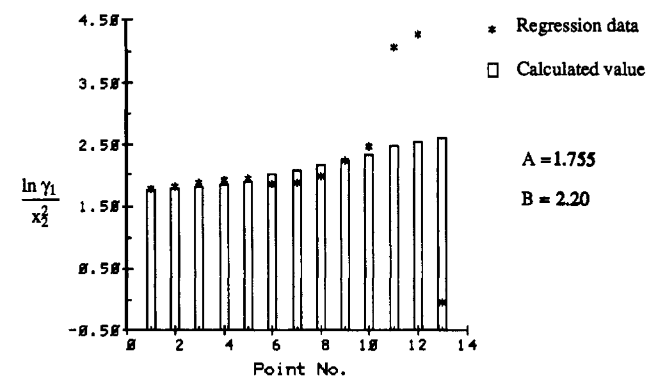

Figure 3. $z_{1,obs}$ and $z_{1,calc}$ when half population linearization with eq 11 is used.

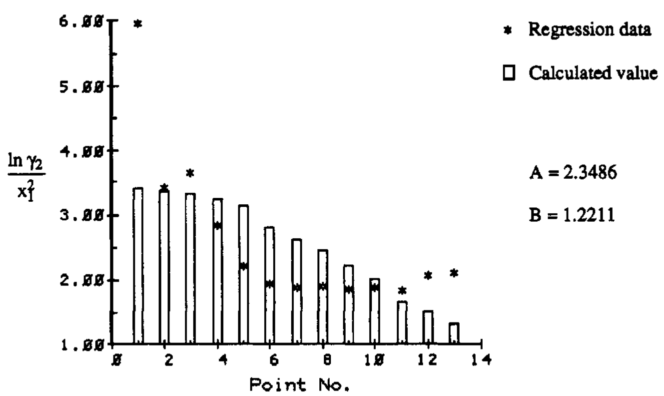

Figure 4. $z_{2,obs}$ and $z_{2,calc}$ when half population linearization with eq 12 is used.

the absolute error is dominant. Using the half or full population linearization transformations, the results indicate large errors. This is shown for the half population case in Figures 3 and 4. Large errors were indeed expected, based on error analysis, because division by very small numbers causes the absolute error to be multiplied many times at certain points when using these transformations. The calculated values of A and B are reasonably close to those calculated by other methods in some cases (see Figure 3, for example), but they are completely wrong in one case (Figure 4).

These results lead to the following conclusions: Linearization which does not involve significant change in the absolute error of the data can give parameter values which are almost as accurate as the parameters obtained by nonlinear regression. Linearization which involves significant increase in the absolute error may give completely incorrect parameter values. Plotting the linearized data in such cases will give a totally unrealistic picture of the error distribution in the measured data.

Table IV. Parameter Values, Sum of Error, and Linear Correlation Coefficient for the Margules Equations

method no.	eq no.	A	B	S^2	R^2
1	4	1.75752	2.11316	0.381871	
2	5	1.7614	2.11	0.382905	
	6	1.5975	2.1436	1.8538	
3	9	1.7752	2.0304	0.608528	0.9928
	10	1.759	2.084	0.412967	0.9955
4	11	1.755	2.20	0.684788	0.07513
	12	2.3486	1.2211	37.3059	0.3914
5	13	1.7157	2.2472	1.07381	0.9951
6	14	1.8557	2.1659	1.4022	0.4685

Data Recovery

The real measure of the accuracy of the constants that have been found is how well they fit the experimental data for γ_1 and γ_2 and thus how far is the sum of squares of errors from the global minimum. Table IV shows the parameter values calculated using the different methods for Margules equations, the respective values of the sum of squares of errors, and the coefficient of determination (R^2).

There are several interesting observations that can be made regarding the results in Table IV. The best values of A and B (with minimal value of S^2) are obtained as expected by nonlinear regression of the full population. Surprisingly accurate results are also obtained when nonlinear regression with half population (γ_1 only) is used. But when the second half of the population (γ_2) is used, the results are much less accurate. Thus, using half population even without linearization can cause substantial error.

The combined effect of linearization and using only half population can be unpredictable. Using eq 10 (with linearization) gives much better results than eq 2 (without linearization), but the use of linearization in eq 12 gives completely wrong results.

The coefficient of determination R^2 (Weisberg, 1980) is a frequently used statistical tool to test the hypothesis that the data points lie on a straight line or can be correlated as a linear function of the coefficients. The value of R^2 is between 0 and 1. R^2 close to 1 indicates very high probability that the data can be well correlated by the linear function. If R^2 is close to 0, this hypothesis should be rejected.

The R^2 value shown for eq 11 in Table IV is very close to 0 ($R^2 = 0.0729$), but the parameters obtained from this equation are very accurate, as can be seen from the small value of S^2. The reason for possible failure of the statistical test in this case is that the test is performed on the transformed data, where the error distribution is completely different from the distribution in the original data.

Figure 5 shows the location of the parameter values calculated using the various methods on a contour plot of S^2 (eq 4) versus A and B. It can be seen that this plot gives excellent indication of the parameter value's accuracy in reproducing the original activity coefficient data. The parameters that are best in reproducing the original data can be expected to predict most accurately new data.

Figure 6 gives a detailed comparison between the experimental data and the most accurate (eq 9) and the least accurate (eq 12) correlations. The best correlation gives a very good representation of the data, while the worst correlation gives completely false results, in particular for low concentrations of either component (high γ_1 or γ_2 values).

Table V shows the parameter values calculated using the different methods for the Van Laar equation, the respective values of the sum of squares of errors, and

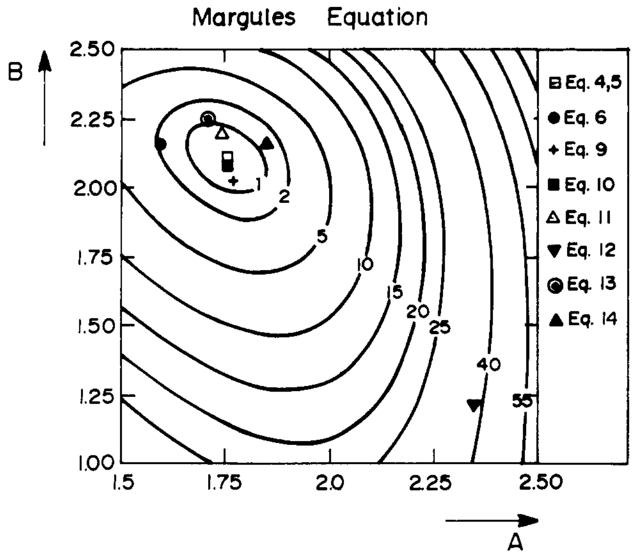

Figure 5. Contour plot of S^2 versus A and B for Margules equation.

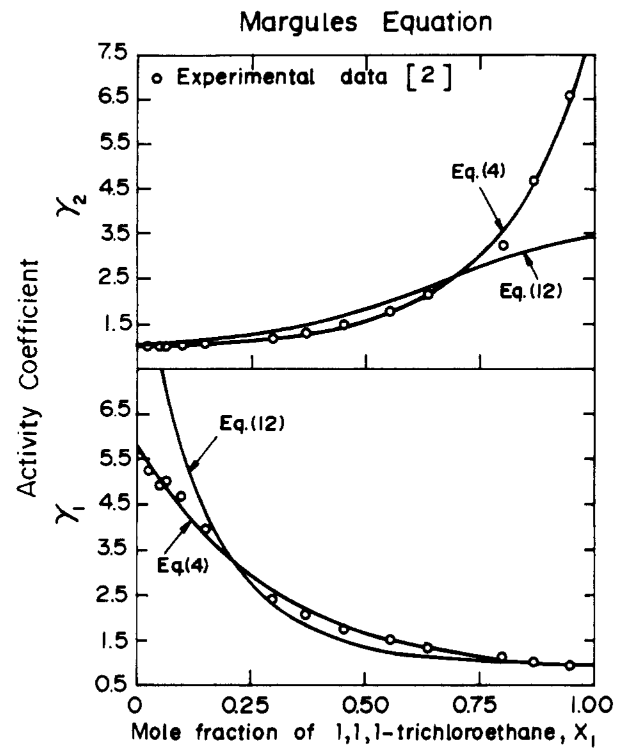

Figure 6. Observed and calculated activity coefficients using the most and least accurate correlation for Margules equation.

Table V. Parameter Values, Sum of Error, and Linear Correlation Coefficient for the Van Laar Equations

method no.	eq no.	A	B	S^2	R^2
1	4	1.76343	2.12554	0.372776	
2	5	1.7667	2.1291	0.37441	
	6	1.6498	2.1579	1.2289	
4	20	1.4112	2.722	14.4018	0.9884
	21	1.791	0.566	49.5998	0.9742
5	23	1.7281	2.3063	1.42504	0.9956
6	24	2.088	2.019	9.34062	0.9953

the linear correlation coefficients. It can be seen that the Van Laar equation is much more sensitive to errors introduced by linearization than the Margules equation. Only the multiple linear regression of full population (eq 23) yields somewhat inaccurate, but acceptable, results. Using nonlinear regression with half population yields very accurate results when using the data of γ_1 and less accurate results when using the data for γ_2. This is very similar to what was observed in the Margules equation. The coefficient of determination R^2, calculated for the cases where it is applicable, obtained a value very close to 1 ($R^2 > 0.97$) in all cases. This should mean accurate correlation, contrary to what the sum of errors indicates. Plotting the most and least accurate correlations versus the data for the Van Laar equation (Figure 7) yields the same conclusion as for the Margules equation.

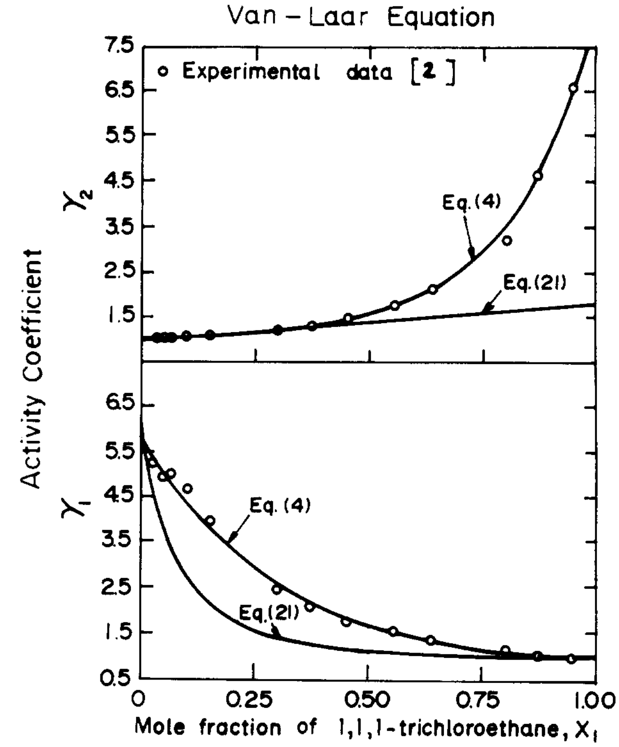

Figure 7. Observed and calculated activity coefficients using the most and least accurate correlation for Van Laar equation.

Conclusions

The objective of this paper was to test the hypothesis that regression of data for the Van Laar and Margules equations, using linearization techniques and/or only half population of the data, can yield parameter values with acceptable accuracy.

It has been demonstrated that this assumption may lead to completely incorrect results. The reason is that any transformation of data may cause significant change in the absolute error distribution which in turn may cause significant differences in the optimal value of the parameters calculated. Linearization can certainly be viewed as data transformation, but the same is true regarding the use of "half population". Even though γ_1 and γ_2 are calculated from the same set of measured data, the calculation itself represents transformation of the data.

Statistical tests performed to check the quality of the fit between the data and the calculated curve can be meaningless if they are performed using transformed data. We have found that locating different sets of parameter values on a contour plot of S^2 versus A and B, as was done in Figure 5, provides the best indication of the accuracy of the parameters.

The most accurate coefficient values can be obtained, undoubtedly, by using nonlinear regression on the full population (eq 4). But as it has been shown, some of the linearization methods, even when using half of the population, can give surprisingly accurate results. So if there is a need to rely solely on such methods, we recommend using several of them and selecting the best coefficients using data similar to the data shown in Tables IV and V.

Acknowledgment

This article was written when one of the authors (M.S.) spent a sabbatical year at the University of Michigan. The hospitality and support of the School of Engineering at the University of Michigan are sincerely appreciated.

Nomenclature

A = Margules or Van Laar equation constant
B = Margules or Van Laar equation constant
g = Gibbs energy
R = universal gas constant
R^2 = coefficient of determination
$R(\,)$ = relative error of a variable
r = the ratio A/B
S^2 = sum of squares of errors
T = temperature, K
V = transformed variable
v = transformed variable
w = transformed variable
x = mole fraction in the liquid phase
y = mole fraction in the vapor phase
z = transformed variable

Greek Letters

γ = activity coefficient
$\delta(\,)$ = absolute error of a variable
ϵ = a small number

Subscripts

1, 2 = subcomponents 1 and 2

Literature Cited

King, M. B. *Phase Equilibrium in Mixtures*; Pergamon Press: Oxford, 1969; pp 316–324.

Kumar, R. K.; Rao, V. M. Vapor-Liquid Equilibria in the Binary Systems Formed by 1-Propanol with 1,2-Dichloroethane 1,1,1-Trichloroethane and 1,1,2,2-Tetrachloroethane at 96.7 kPa. *Fluid Phase Equilib.* **1991**, *70*, 19–32.

Walas, S. M. *Phase Equilibria in Chemical Engineering*; Butterworth Publishers: London, 1985.

Weisberg, S. *Applied Linear Regression*; John Wiley & Sons: New York, 1980, pp 18, 19, 48, 49.

Wisniak, J. Pitfalls in Fitting Binary Vapor-Liquid Equilibria Data with Two-Parameter Equations. *Fluid Phase Equilib.* **1993**, in press.

Received for review February 1, 1993
Revised manuscript received June 10, 1993
Accepted July 12, 1993[®]

[®] Abstract published in *Advance ACS Abstracts,* October 1, 1993.