LIQUID–VAPOUR EQUILIBRIUM LVII.*
MULTICOMPONENT SYSTEMS

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A thermodynamically consistent procedure is proposed based on successive excess free energy expansions which leads to flexible equations expressing the concentration dependence of activity coefficients. In first approximation, equations of two-suffix type are deduced which describe multicomponent systems by means of binary constants free of restraining conditions and which lead, after introducing simplifying assumptions, to Renon–Prausnitz NRTL, Black, Wohl–van Laar and Wohl–Margules equations.

Vapour–liquid equilibrium data are usually reduced to thermodynamic functions (excess free energy of mixing and activity coefficients) and adjusted to give meaningful correlations and generalizations. Many equations have been proposed for expressing the composition and temperature dependence of the activity coefficients. Well known are the Margules[1], van Laar[2] (in Carlson–Colburn modification[3]), Scatchard–Hamer[4] and Redlich–Kister[5] types. Wohl[6,7] showed that a series expansion of exess free energy in effective volume fractions leads after introducing certain simplifying conditions to all these equations.

Wohl deduced as first approximation equations of "two-suffix type" which describe behaviour of mixture by means of binary constants only. The most simple symmetrical or regular equation characterizes the M-component system with the aid of $0.5M(M-1)$ constants which all can be evaluated from binary experimental data (*i.e.* binary system is described by means of one constant, ternary system by means of three constants *etc.*).

Unfortunately, the practical experience shows that regular equations are not sufficient to characterize quantitatively the majority of binary systems and to predict the behaviour of three- and more-component systems. Wohl's nonsymmetrical two-suffix type, which is fully equivalent to van Laar equation, describes the M-component system by means of $M(M-1)$ constants (*i.e.* binary system be means of two constants, ternary system by means of six constants *etc.*). Practical experience shows that van Laar equation is flexible and able to characterize many (but not all) binary systems[8,9]. Its ability to predict behaviour of three- and more-component systems from binary data can be estimated as semiquantitative.

It follows from the preceeding discussion that simple two-suffix equations are not fully convenient for the correlation of binary data and not sufficient for the quantitative prediction of behaviour of multicomponent systems by means of binary constants. For this reason, new procedures have been looked for to improve the flexibility and the prediction ability of the correlation equations. Wohl[6,7], Redlich–Kister[5] and Hála[10] introduced the third and higher terms from

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excess free energy series expansion and deduced equations of three-, four- and higrher-suffix type. Many but not all constants of these equations can be evaluated from binary data. Ternary, eventually quarternary experimental data are necessary for the quantitative characterization of multicomponent systems (details see[11]). Three- and four-suffix procedure is fully justified from the phenomenological point of view. The resulting equations are thermodynamically consistent and their flexibility is better than in the case of two-suffix types. From the molecular point of view, the justification of three- and four-suffix equations is questionable. The statistical theories of liquid mixtures are based on binary molecular interactions[12]. This fact leads to the logical deduction that behaviour of multicomponent systems should be (at least in many cases) predicted from the known behaviour of binary mixtures. For this reason new types of more flexible equations (with binary constants only) have been looked for. In 1964 Wilson[13] derived an equation using a procedure which is in certain sence generalization of Flory–Huggins theory. His equation has two adjustable constants per binary system and its flexibility and ability to predict the behaviour of three- and more-component systems is very good, as it has been practically shown[14]. It must be mentioned that Wilson equation is not applicable to partially miscible liquid mixtures. To overcome this difficulty, Renon and Prausnitz[15–18] combined Wilson's local mole fraction concept with Scott's two liquid theory of mixture[19] and derived NRTL equation (Non Random-Mixing Two-Liquid) which has three adjustable parameters per binary system. NRTL equation is very flexible, can be used for systems with limited miscibility and extended to mixtures of more than two components. Extensive practical tests have shown[15,20,21] that its flexibility is excellent and its ability to predict behaviour of multicomponent systems good.

It must be, however, taken into account that certain restraining conditions must be fulfilled between constants of both van Laar and Wilson and NRTL equation in three- and more-component systems if we wish to be consistent with the theoretical models[22]. This fact complicates the practical use of the just named equations.

It is shown in this contribution that a general procedure can be developped based on superponed excess free energy expansions which leads to flexible equations expressing the concentration dependence of activity coefficients.

Excess Free Energy of Superponed Type

As first approximation, equations of "two-suffix successive type" can be deduced which describe multicomponent system by means of binary constants free of restraining conditions. Let us suppose that excess free energy of the M-component system can be expressed as a set of particular contributions

$$\Delta G^E \big/ RT = \sum_{r=1}^{M} Q_r , \qquad (1)$$

where ΔG^E is excess free energy of mixing, T absolute temperature and R gas constant. Each term of the sum, Q_r, is function of composition, temperature, pressure and total number of mol es (Q_r is extensive property of the system). Its concentration dependence can be expressed with the aid of specific concentration variables, $(z_i)_r$, defined by

$$(z_i)_r = n_i(q_i)_r \big/ \sum_{j=1}^{M} n_j(q_j)_r , \qquad (2)$$

where n_i, n_j are numbers of mol of component, i, j and $(q_i)_r$, $(q_j)_r$ constants characteristic for the component i, j in the particular term Q_r. The sum of effective fractions of all components is equal to unity

$$\sum_{j=1}^{M} (z_j)_r = 1 . \tag{3}$$

Let us now define an intensive excess function Q_r' and express its concentration dependence by means of series expansion. Since in the limit for $(z_i)_r = 1$; $(i = 1, 2, \ldots \ldots, M)$ Q_r' is equal to zero, all single index terms of the series vanish and the dependence on composition can be written in the form

$$Q_r' = Q_r / \sum_{j=1}^{M} n_j (q_j)_r = \sum_{ij} (z_i)_r (z_j)_r (a_{ij})_r + \ldots, \tag{4}$$

where $(a_{ij})_r$ are constants and the summation gives the sum of the products of the concentration variables $(z_i)_r (z_j)_r$ of all pair dissimilar constituents that can be chosen from the given M-component system, where each product is multiplied by a constant $(a_{ij})_r$. For example in a binary system

$$\sum_{ij} (z_i)_r (z_j)_r (a_{ij})_r = (z_1)_r (z_2)_r (a_{12})_r + (z_2)_r (z_1)_r (a_{21})_r = 2(z_1)_r (z_2)_r (a_{12})_r , \tag{5}$$

since $(a_{12})_r = (a_{21})_r$. Similarly for a ternary system

$$\begin{aligned}
\sum_{ij} (z_i)_r (z_j)_r (a_{ij})_r &= (z_1)_r (z_2)_r (a_{12})_r + (z_2)_r (z_1)_r (a_{21})_r + \\
&+ (z_1)_r (z_3)_r (a_{13})_r + (z_3)_r (z_1)_r (a_{31})_r + (z_2)_r (z_3)_r (a_{23})_r + \\
&+ (z_3)_r (z_2)_r (a_{32})_r = 2(z_1)_r (z_2)_r (a_{12})_r + \\
&+ 2(z_1)_r (z_3)_r (a_{13})_r + 2(z_2)_r (z_3)_r (a_{23})_r .
\end{aligned} \tag{6}$$

Binary System

We consider as a first example the derivation of two-suffix equation for a binary system. According to Eqs (*1*) and (*4*)

$$\Delta G^E / RT = Q_1 + Q_2 = [n_1(q_2)_1 + n_2(q_2)_1] (z_1)_1 (z_2)_1 \, 2(a_{12})_1 + \\ + [n_1(q_1)_2 + n_2(q_2)_2] (z_1)_2 (z_2)_2 \, 2(a_{12})_2 . \tag{7}$$

If we introduce new constants

$$\begin{aligned}
(A_{12})_1 &= 2(a_{12})_1 (q_1)_1 , \qquad (A_{21})_1 = 2(a_{12})_1 (q_2)_1 , \\
(A_{12})_2 &= 2(a_{12})_2 (q_1)_2 , \qquad (A_{21})_2 = 2(a_{12})_2 (q_2)_2 ,
\end{aligned} \tag{8}$$

we can write

$$\Delta G^E/RT = n_1 (z_2)_1 (A_{12})_1 + n_2 (z_1)_2 (A_{21})_2 . \tag{9}$$

The relations giving the dependence of the activity coefficients on the composition of the solution are obtained by partial differentiation of the excess free energy according to n_1 and n_2

$$\ln \gamma_1 = \left[\frac{\partial}{\partial n_1} \left(\frac{\Delta G^E}{RT} \right) \right]_{T, P, n_2} = (A_{12})_1 (z_2)_1^2 + (A_{12})_2 (z_2)_2^2 ,$$

$$\ln \gamma_2 = \left[\frac{\partial}{\partial n_2} \left(\frac{\Delta G^E}{RT} \right) \right]_{T, P, n_1} = (A_{21})_1 (z_1)_1^2 + (A_{21})_2 (z_1)_2^2 . \tag{10}$$

Equations (9) and (10) contain four constants, *i.e.* $(A_{12})_1$, $(A_{21})_1$, $(A_{12})_2$, $(A_{21})_2$ which must be evaluated from experimental measurements and which are connected with the limiting values of the natural logarithm of the activity coefficient

$$\lim_{x_1 \to 0} \ln \gamma_1 = (A_{12})_1 + (A_{12})_2 , \tag{11}$$

$$\lim_{x_2 \to 0} \ln \gamma_2 = (A_{21})_1 + (A_{21})_2 .$$

By introducing various simplifying assumptions equations (9) and (10) are converted to relations derived previously by various authors. Thus, if we introduce

$$\tau_{21} = (A_{12})_1 , \qquad \tau_{12} = (A_{21})_2 \tag{12}$$
$$G_{21} = (A_{21})_1/(A_{12})_1 , \qquad G_{12} = (A_{12})_2/(A_{21})_2 ,$$

equations (9) and (10) take the form of Renon–Prausnitz NRTL type[15,16] which constains three freely adjustable parameters. The fourth is given by means of equation

$$\frac{(A_{12})_1}{(A_{21})_2} = \frac{\ln \left[(A_{12})_1/(A_{21})_1 \right]}{\ln \left[(A_{21})_2/(A_{12})_2 \right]} . \tag{13}$$

If we assume that

$$(A_{12})_2 = (A_{21})_2 = A , \tag{14}$$

relations (9) and (10) degenerate into Black equation[23]. On the assumption that

$$(A_{12})_2 = (A_{21})_2 = 0 , \tag{15}$$

equations (9) and (10) take the form of Wohl twosuffix type[6,7] which is fully equivalent to van Laar equation (in Carlson–Colburn modification[3]). Finally, if

$$(A_{12})_1 = (A_{21})_1 = A,$$
$$(A_{12})_2 = (A_{21})_2 = 0,$$
(16)

equations further simplify to Wohl–Margules symmetrical form[7,13].

Ternary and Multicomponent Systems

Equation (1) has in the ternary system the following form

$$\Delta G^{\mathrm{E}}/RT = \sum_{r=i}^{k} Q_r = Q_i + Q_j + Q_k,$$
(17)

where

$$Q_r = [n_i(q_i)_r + n_j(q_j)_r + n_k(q_k)_r] \cdot$$
$$\cdot [(z_i)_r (z_j)_r 2(a_{ij})_r + (z_i)_r (z_k)_r 2(a_{ik})_r + (z_j)_r (z_k)_r 2(a_{jk})_r].$$
(18)

If we introduce new constants

$$(A_{ij})_r = 2(a_{ij})_r (q_i)_r, \qquad (A_{ji})_r = 2(a_{ij})_r (q_j)_r,$$
$$(A_{ik})_r = 2(a_{ik})\cdot(q_i)_r, \qquad (A_{ki})_r = 2(a_{ik})_r (q_k)_r,$$
$$(A_{jk})_r = 2(a_{jk})_r (q_j)_r, \qquad (A_{kj})_r = 2(a_{jk})_r (q_k)_r,$$
(19)

we can rearrange Eqs (18) and (1) and write

$$Q_r = n_i(z_j)_r (A_{ij})_r + n_i(z_k)_r (A_{ik})_r + n_j(z_k)_r (A_{jk})_r,$$
(20)

$$\Delta G^{\mathrm{E}}/RT = n_i(z_j)_i (A_{ij})_i + n_i(z_k)_i (A_{ik})_i + n_j(z_k)_i (A_{jk})_i +$$
$$+ n_j(z_k)_j (A_{jk})_j + n_j(z_i)_j (A_{ji})_j + n_k(z_i)_j (A_{ji})_j +$$
$$+ n_k(z_i)_k (A_{ki})_k + n_k(z_j)_k (A_{kj})_k + n_i(z_j)_k (A_{ik})_k.$$
(21)

It must be mentioned that five constants defined for each term Q_r by Eq. (19) are free. The sixth fulfils the restraining condition

$$\frac{(A_{kj})_r}{(A_{jk})_r} = \frac{(A_{ki})_r}{(A_{ik})_r} \left| \frac{(A_{ji})_r}{(A_{ij})_r} \right. .$$
(22)

It follows from Eqs (19), (21) and (22) that the general form of the two-suffix equation has the same disadvantage as equations proposed by Wohl (van Laar), Black and Renon–Prausnitz NRTL, *i.e.*, that constants evaluated from binary data can not be used directly to the characterization of three-component systems. The entire adjustment must be made in such a way that restraining condition (22) is fulfilled. In this situation it is, however, possible to take into account the very high flexibility of the two-suffix superponed equation (21). If we suppose that the constant $(A_{jk})_r$ is very small, $(A_{kj})_r$ must be also very small, as it follows from Eq. (22)

$$(A_{jk})_r \ \to \ 0 : \quad (A_{kj})_r \ \to \ 0 , \tag{23}$$

and the three-component system is characterized by means of twelve binary constants which are fully free and which can be evaluated from binary experimental data. Equation (21) degenerates in conection with condition (23) into the form

$$\Delta G^E / RT = n_i(z_j)_i \, (A_{ij})_i + n_i(z_k)_i \, (A_{ik})_i + n_j(z_k)_j \, (A_{jk})_j +$$
$$+ \, n_j(z_i)_j \, (A_{ji})_j + n_k(z_i)_k \, (A_{ki})_k + n_k(z_j)_k \, (A_{kj})_k \tag{24}$$

and the dependence of the activity coefficient of component i in a ternary system can be expressed as follows:

$$\ln \gamma_i = \left[1 - (z_i)_i \right] \left[(z_j)_i \, (A_{ij})_i + (z_k)_i \, (A_{ik})_i \right] +$$
$$+ \, (z_j)_j [(A_{ij})_j / (A_{ji})_j] \left[(A_{ji})_j - (z_i)_j \, (A_{ij})_j - (z_k)_j \, (A_{jk})_j \right] +$$
$$+ \, (z_k)_k [(A_{ik})_k / (A_{ki})_k] \left[(A_{ki}) - (z_i)_k \, (A_{ki})_k - (z_j)_k \, (A_{kj})_k \right] . \tag{25}$$

Extension of the procedure to M-component system is evident. Each term, Q_r, in Eq. (1) can be written in the form

$$Q_r = \sum_{i=1}^{M-1} n_i \big[\sum_{j>i} (z_j)_r \, 2(a_{ij})_r \, (q_i)_r \big] . \tag{26}$$

If we introduce new constants

$$(A_{ij})_r = 2(a_{ij})_r \, (q_i)_r , \qquad (A_{ji})_r = 2(a_{ij})_r \, (q_j)_r , \tag{27}$$

we can rearrange Eq. (26) and write

$$Q_r = \sum_{i=1}^{M-1} n_i \big[\sum_{j>i} (z_j)_r \, (A_{ij})_r \big] . \tag{28}$$

The contribution of the term Q_r to the activity coefficient of component it is obtained by partial differentiation of Eq. (28) according to n_i

$$\left(\partial Q_r/\partial n_i\right)_{T,P,n_{j\ne r}} = \sum_{j>i}\left(z_j\right)_r\left(A_{ij}\right)_r - \sum_{j=1}^{M-1}\left(z_j\right)_r\left[\sum_{k>j}\left(z_k\right)_r\left(A_{jk}\right)_r\left(A_{ij}\right)_r/\left(A_{ji}\right)_r\right]. \quad (29)$$

The discussion of restraining conditions (22) is similar to the preceeding ternary system. If we suppose

$$\left(A_{jk}\right)_r \to 0 \; ; \qquad \left(A_{kj}\right)_r \to 0 \, , \quad (23)$$

the last terms in Eq. (29) can be neglected and the final equations take the form

$$\Delta G^E/RT = \sum_{i=1}^{M} n_i\left(\sum_{j\ne i}\left(z_j\right)_i\left(A_{ij}\right)_i\right), \quad (30)$$

$$\ln \gamma_i = \left[\frac{\partial\left(\Delta G^E/RT\right)}{\partial n_i}\right]_{T,P,\,n_{j\ne i}} = \left[1 - \left(z_i\right)_i\right]\left[\sum_{j\ne i}\left(z_j\right)_i\left(A_{ij}\right)_i\right] +$$
$$+ \sum_{j\ne i}\left(z_j\right)_i\frac{\left(A_{ij}\right)_j}{\left(A_{ji}\right)_j}\left[\left(A_{ji}\right)_j - \left(\sum_{i\ne j}\left(z_i\right)_j\left(A_{ji}\right)_j\right)\right]. \quad (31)$$

Equations (30) and (31) characterize the M-component system by means of $4M$ constants which are free and which can be evaluated from binary experimental data.

The two-suffix equation (30) can be converted to other useful types by introducing certain simplifying assumptions. Thus, if we introduce

$$\frac{\left(A_{ij}\right)_r}{\left(A_{ji}\right)_s} = \frac{\ln\left[\left(A_{ij}\right)_r/\left(A_{ji}\right)_r\right]}{\ln\left[\left(A_{ji}\right)_s/\left(A_{ij}\right)_s\right]}, \qquad \begin{bmatrix} i < j \\ r < s \end{bmatrix}, \quad (32)$$

and define new constants

$$\tau_{ji} = \left(A_{ij}\right)_r : \qquad \tau_{ij} = \left(A_{ji}\right)_s \, ,$$
$$G_{ji} = \left(A_{ji}\right)_r/\left(A_{ij}\right)_r , \qquad G_{ij} = \left(A_{ij}\right)_s/\left(A_{ji}\right)_s , \quad (33)$$

equations (30) and (31) take the form of NRTL multicomponent type with *fully free binary constants*.

It must be mentioned that practically all authors who have predicted multicomponent system's behaviour from binary data by means of NRTL equation[15-17,20] did not take into account the restraining conditions[22]

$$\tau_{jk} - \tau_{kj} = \tau_{ik} - \tau_{ki} - \left(\tau_{ij} - \tau_{ji}\right) \quad (34)$$

and correlated their data in fact by means of equation (31).

If we assume that

$$\left(A_{ij}\right)_2 = \left(A_{ji}\right)_2 \, ,$$
$$\left(A_{ij}\right)_{r\ne1,2} = \left(A_{ji}\right)_{r\ne1,2} = 0 \, , \quad (35)$$

relation degenerates into Black equation[23]. On the assumption that

$$(A_{ij})_{r \neq 1} = (A_{ji})_{r \neq 1} = 0,$$ (36)

equation (1) takes the form of Wohl–van Laar two-suffix type[6,7]. Finally, if

$$(A_{ij})_1 = (A_{ji})_1 = (A_{ij}),$$
$$(A_{ij})_{r \neq 1} = (A_{ji})_{r \neq 1} = 0,$$ (37)

equation (1) further simply to Wohl-Margules symmetrical form[6,7].

ADDENDUM

Palmer D. A. and Smith B. D. (Ind. Eng. Chem. Process Des. Develop. *11*, 114 (1972)) published recently new two-parameter local-composition equation capable of correlating systems with partial miscibility

$$\frac{\Delta G^E}{RT} = x_1 x_2 \left(\frac{\Theta_{21} \tau_{21}}{x_1 + x_2 \Theta_{21}} + \frac{\Theta_{12} \tau_{12}}{x_2 + x_1 \tau_{12}} \right),$$

where constants $\tau_{12}, \tau_{21}, \Theta_{12}, \Theta_{21}$ are defined by means of equations

$$\tau_{12} = \ln(P'_{12}/P_{22}), \quad \tau_{21} = \ln(P'_{21}/P'_{11}),$$

$$P'_{12} = P'_{21},$$

$$\Theta_{12} = \exp \left(\frac{\Delta H^V_{12} - \Delta H^V_{22}}{RT} + \ln \frac{P'_{12}}{P'_{22}} \right),$$

$$\Theta_{21} = \exp \left(\frac{\Delta H^V_{12} - \Delta H^V_{11}}{RT} + \ln \frac{P'_{21}}{P'_{11}} \right),$$

P'_{11}, P'_{22} denote vapour pressure of pure component 1, 2, $\Delta H^V_{11}, \Delta H^V_{22}$ heat of vaporization of pure component 1, 2 and P'_{12} and $\Delta H'_{12}$ are adjusting parameters.

It can be shown that Palmer–Smith two-parameter equation can be deduced from equation (9) if we introduce

$$(A_{12})_1 = \tau_{21} = \ln \frac{P'_{12}}{P'_{11}},$$

$$(A_{21})_1 = \tau_{21} \Theta_{21} = \left(\ln \frac{P'_{12}}{P'_{11}} \right) \exp \left(\frac{\Delta H^V_{12} - \Delta H^V_{11}}{RT} + \ln \frac{P'_{12}}{P'_{11}} \right),$$

$$(A_{12})_2 = \tau_{12} \Theta_{12} = \left(\ln \frac{P'_{12}}{P'_{22}} \right) \exp \left(\frac{\Delta H^V_{12} - \Delta H^V_{22}}{RT} + \ln \frac{P'_{12}}{P'_{22}} \right),$$

$$(A_{21})_2 = \tau_{12} = \ln \frac{P'_{12}}{P'_{22}}.$$

Two constants of equation (9) are then free and two are related by restraining conditions

$$(A_{21})_2 = (A_{12})_1 + \ln (P'_{11}/P'_{22}),$$

$$\frac{(A_{12})_1 (A_{12})_2}{(A_{21})_1 (A_{21})_2} = \exp \left(\frac{\Delta H_{11}^Y - \Delta H_{22}^Y}{RT} + \ln \frac{P'_{11}}{P'_{22}} \right).$$

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Translated by the author.