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A GENERAL SOLUTION MODEL FOR PREDICTING TERNARY THERMODYNAMIC PROPERTIES

Kuo-Chih Chou
University of Science and Technology Beijing
Beijing, People's Republic of China

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ABSTRACT All current models used to predict ternary thermodynamic properties from binaries improperly assume that the selected binary compositions in a ternary model are independent of ternary considered. As a result, it will require human interference for selecting models and arranging components to three apexes of a triangle, and will lead to an unacceptable result in some limiting case. A reasonable assumption should be that the selected binary compositions are closely related to the ternary itself. On the basis of this idea, a completely new model has been suggested. It seems that this new model is more reasonable theoretically from consideration, more reliable in practical application and more realistic to use in computerized thermodynamic and phase diagram calculation.

I. Introduction

There is a considerable need of thermodynamic properties and phase diagrams, especially for the ternary and multicomponent systems where there is a shortage of thermodynamic data in these systems. Unfortunately, this problem is not easy to be solved experimentally due to the complexity of experiments in a multicomponent system. As a result, the calculation of thermodynamic properties and phase diagrams have become increasingly important. It may be expected that most of ternary and multicomponent thermodynamic data will come from the calculation rather than experiments directly.

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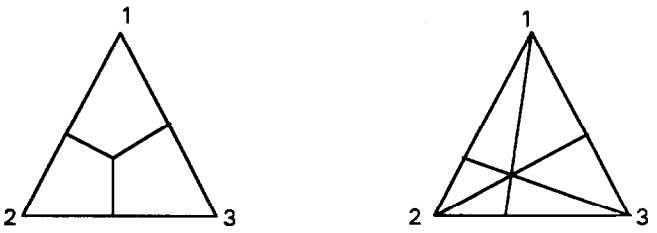
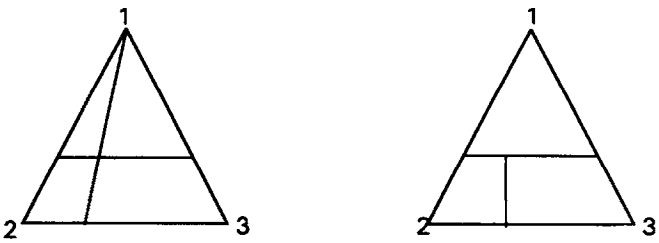
Predicting thermodynamic properties for ternary and multicomponent systems from binary ones is the most attractive and powerful method among all theoretical methods because it is simple and effective and only requires information that is easy to obtain. It has already been widely used in the calculation of phase diagrams and estimation of thermodynamic properties for ternary and multicomponent systems in the past decades. The core of this numerical method can be related to a choice of the model that can express the ternary thermodynamic properties in terms of its three binaries each with an assigned binary composition.



(a) Two kinds of symmetrical models



(b) Two kinds of asymmetrical models
Fig.1. Symmetrical and Asymmetrical Models

According to the method of selecting the binary composition, Hillert[1] has classified these models into two categories: symmetrical and asymmetrical. The Kohler[2], Colinet[3] and Muggianu[4] models belong to the symmetric models whereas the Bonnier[5], Toop[6] and Hillert[1] models belong to asymmetric ones (Fig.1). From 1960 to 1986 around 7 or 8 models have been published. Ansara[7,8] and Hillert [1,9] have given a good review, summary and discussion for these models in early 1970 and 1980. Based on their enlightening summary and analysis, the author and his group have found that, all these models can be summarized as [10-18]

$$\Delta G^{E} = W_{12}\Delta G^{E}_{12} + W_{31}\Delta G^{E}_{31} + W_{23}\Delta G^{E}_{23} \qquad (1)$$

where ΔG^{E} and ΔG^{E}_{ij} represent the ternary and binary Gibbs energies of mixing respectively and W_{ij} indicates the weight probability, and further found the basic relation connecting the selected binary composition ($X_{i(ij)}, X_{j(ij)}$) and weight probability as[18]

$$W_{ij} = \frac{x_i x_j}{X_{i(ij)} X_{j(ij)}} \qquad (2)$$

where x_i and x_j represent the mole fraction of components in a ternary system. Since then, we have developed dozens and dozens of models between 1987 to 1989 [10-18]. A systematic summary of these models was given by Chou and Chang[18], in which all kinds of models including the above typical models and several dozens of other possible potential models as well as their corresponding calculation formulae are discussed. It is worthy mentioning that, the so called integration model developed by Chen et al.[17] is also a special kind of model that has successfully been applied to some practical systems. In order to distinguish so many models, a new nomenclature system for these models is suggested[18]. As a matter of fact, our method can give an infinite number of models since the function "δ " used in our formula can be any value, each δ corresponds to a W_{ij} and a new model[18]. Besides, two points can be a model, three points, four points...etc. also can construct a model[18] and finally integration in a certain concentration range also can be a model as we did before[17]. In this way, there will be no ending. Therefore, it is already meaningless to continue to create new models in this way. We should pay our attention in checking all of these models, comparing with experimental data, analyzing what problems they have and how to improve them. It is the object of this paper.

II. The Problem of Current Models

In the reference[18], we have given a definition of symmetric and asymmetric models, that is, "a model shall be referred to as a symmetric model if the ternary Gibbs free energy of mixing can be expressed in terms of its three binary Gibbs free energies of mixing at the same kind of selected composition and the same kind of assigned probability weight for all three binaries, all other cases shall be deemed to be asymmetric." Figure 1 shows two kinds of symmetric model and two kinds of asymmetric model as well as their corresponding selected composition points for binaries respectively. It may be seen from Figure 1 that, with regard to an asymmetrical model like Toop's or Hillert's, the three selected binary compositions are different in three binaries, that means, a different arrangement of three components to three apexes of triangle will lead to a different result of ternary Gibbs free energy of mixing. Ansara[7] and other investigators have already noted this fact in their papers. It has been recommended by some researchers that one should assign three components to three apexes of a triangle in terms of the characteristics of components such as valence, structure and the element position in the periodical table...,etc. For example, for the AgCl-$MgCl_2$-$CaCl_2$ ternary system, it is suggested one should choose corner "1" for AgCl and "2","3" for $MgCl_2$, $CaCl_2$ to construct a Toop model because Mg^{++}, Ca^{++} are both divalent. However, evidently this kind of selection is completely man-made and sometimes may not work. Besides, the procedure will become very complicated as the number of components gets larger and larger. It will be discussed in our next paper concerning a multicomponent model.

The symmetric model could avoid this trouble since the selections for three binary compositions are made in exactly the same way. However, they lead to other problems. It is well known that, any reasonable model should be able to reduce to its limiting form if the limiting conditions are met. For example, a good ternary solution model should be simplified to a binary solution model if the characteristics of the third component is identical to that of the second one, since, in fact, there are only two components "1" and "2" in the system. Unfortunately, the symmetric model is unable to meet this basic requirement[1]. It may be seen

from Figure 1 that, for any one of the symmetric models (Kolher or Muggianu), if the third component is identical to the second component, the two selected compositions corresponding to "1-2" and "1-3" respectively do not coincide and the selected composition of component "1" in the binaries "12" and "31" is not equal to x_1, i.e., $X_1 \neq x_1$(here the capital X denotes binary composition and lower case x ternary composition), that means, this binary system "1-2" ("3" and "2" are identical now) still has two compositions to be selected. Apparently, it is unreasonable.

In brief, both symmetrical and asymmetrical models have their inherent problem. For a symmetric model, it can not reduce to a binary system even if two out of three components are identical. It is certainly unacceptable at least from the theoretical point of view. For an asymmetrical model, there will be an undetermined factor about how to distribute three components into three apexes of a triangle.

In addition, when a model is applied, first one may ask what kind of model should be used, symmetrical or asymmetrical, second, if an asymmetrical model is selected, one may further ask how to assign three components to three apexes in a triangle. All of these selections are man-made and computer doesn't know how to do that. This problem will become very serious when dealing multicomponent system with a large number of components. Obviously, these problems can not be solved in terms of current models based on a symmetrical or asymmetrical type due to its inherent defects. Thus we have to get rid of the traditional way and try some completely new approach to solve these problems.

III. A New General Model

At first, let us define a quantity η_I that is called the "deviation sum of squares",

$$\eta_{\mathrm{I}} = \int_0^1 (\Delta G^{\mathrm{E}}_{12} - \Delta G^{\mathrm{E}}_{13})^2 \mathrm{d}X_1 \tag{3}$$

where ΔG^E_{12} and ΔG^E_{13} represents the excess Gibbs energy of binary solution "12" and "13" respectively, X_1 indicates the mole fraction of component 1 in "12" or "31" binary solutions. Similarly one has

$$\eta_{\mathrm{II}} = \int_0^1 (\Delta G^{\mathrm{E}}_{21} - \Delta G^{\mathrm{E}}_{23})^2 \mathrm{d}X_2 \tag{4}$$

$$\eta_{\mathrm{III}} = \int_0^1 (\Delta G^{\mathrm{E}}_{31} - \Delta G^{\mathrm{E}}_{32})^2 \mathrm{d}X_3 \tag{5}$$

Obviously, if the component "3" is similar to the component "2" thermodynamically, the value of η_I should approach zero otherwise a positive non zero value of η_I will be expected. Now let us introduce another quantity, the so called "similarity coefficient" which is defined as

$$\xi_{12} = \frac{\eta_{\mathrm{I}}}{\eta_{\mathrm{I}} + \eta_{\mathrm{II}}} \tag{6}$$

If the third component is similar to the second one, $\eta_I = 0$, thus $\xi_{12} = 0$, and if the third one is similar to the first one, $\eta_{II} = 0, \xi_{12} = 1$. Therefore from the ξ_{12} value one can judge if the third component is more similar to the component one or two. Analogously, we have

$$\xi_{31} = \frac{\eta_{III}}{\eta_{III} + \eta_{I}} \tag{7}$$

$$\xi_{23} = \frac{\eta_{II}}{\eta_{II} + \eta_{III}} \tag{8}$$

On the basis of the above definitions, the following binary composition will be selected for this new model,

$$X_{1(12)} = x_1 + x_3\xi_{12} \tag{9}$$

$$X_{3(31)} = x_3 + x_2\xi_{31} \tag{10}$$

$$X_{2(23)} = x_2 + x_1\xi_{23} \tag{11}$$

where $X_{i(ij)}$ denotes the mole fraction of component "i" in the "ij" binary system. Since the value of ξ will range from 0 to 1, so the range of $X_{1(12)}$ varies from x_1 to $(x_1 + x_3)$ or from point "a" to point "b", as shown in Fig.2.(please note again, the capital X used here indicating binary composition is different from the lower case x which is used to represent the ternary composition).

Substituting Eqs.(9) to (11) into Eq.(2) and combining with Eq.(1), one will obtain the expression of the ternary excess Gibbs energy for this new model, that is

$$\Delta G^{E} = \frac{x_1 x_2}{(x_1 + \xi_{12}x_3)(x_2 + (1-\xi_{12})x_3)}\Delta G^{E}_{12} + \frac{x_3 x_1}{(x_3 + \xi_{31}x_2)(x_1 + (1-\xi_{31})x_2)}\Delta G^{E}_{31} + \frac{x_2 x_3}{(x_2 + \xi_{23}x_1)(x_3 + (1-\xi_{23})x_1)}\Delta G^{E}_{23} \tag{12}$$

This is the formula of the new model used to calculate the Gibbs free energy of mixing for a ternary system.

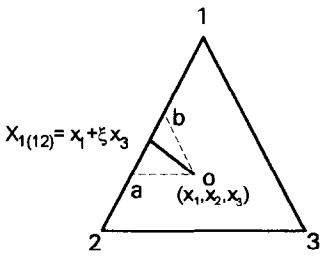


Fig.2. The relation between ternary and binary compositions for this new model

IV. The Relationship Between Three Similarity Coefficients

Three similarity coefficients for three binaries are not independent. Combining Eqs.(6) to (8) and eliminating $\eta_I, \eta_{II}, \eta_{III}$ yield

$$(1-\xi_{12})(1-\xi_{31})(1-\xi_{23}) = \xi_{12}\xi_{31}\xi_{23} \tag{13}$$

The above equation provides the mechanism for us to calculate the third similarity coefficient from the other two known coefficients. From the above equation, it may be seen that, if any one of the three similarity coefficients approaches zero, then there must be one similarity coefficient which approaches unity. The same conclusion is true for the converse situation, i.e. if any one similarity coefficient approaches unity, there must be a coefficient also close to zero. The above situation represents the common feature of Bonnier, Toop and Hillert models. From the above equation, one may also find that, if two out of three similarity coefficients are equal to 0.5, then the third one must be equal to 0.5 too. This case represents the Muggianu model.

V. Discussions

1). This new model can reduce to various kinds of simple limiting form if the different limiting conditions are fulfilled. For example, if three binaries are all ideal solution, i.e. $\Delta G_{12}^E = \Delta G_{31}^E = \Delta G_{23}^E = 0$, then the ternary solution becomes an ideal ternary solution since $\Delta G^E = 0$. When three binaries are regular solutions, i.e. $\Delta G_{12}^E = A_{12}^0 X_1 X_2$, $\Delta G_{31}^E = A_{31}^0 X_3 X_1$, $\Delta G_{23}^E = A_{23}^0 X_2 X_3$, substituting these expressions into Eq.(1) and combining with equation (2), the ternary Gibbs free energy of mixing will be

$$\Delta G^E = A_{12}^0 x_1 x_2 + A_{31}^0 x_3 x_1 + A_{23}^0 x_2 x_3 \tag{14}$$

This is a ternary regular solution. These characteristics are the same as all other models presented before.

2) This new model differs from all other models on its special selection of binary compositions which are closely related to the ternary system considered. For instance, the selection of binary composition in "12" system will depend on the characteristics of systems "31" and "23", as shown in Eqs.(6) and (9). If the characteristics of the system "12" is similar to the system "31" but deviating from "23", i.e. $\eta_I = 0$ and η_{II} with a positive non zero value, thus according to Eqs.(6) and (9), $\xi_{12} = 0$, $X_{1(12)} = x_1$ that is the smallest limiting value of X_1 that one can have. On the contrary, if system "12" is similar to "23" but deviating from "31", one will get a larger $X_{1(12)}$, the largest possible limiting value will be (x1 + x3) due to $\xi_{12} = 1$. However, all current models select the binary compositions in three binaries in an unchangeable manner.

3).When the third component "3" is exactly the same as component "2" in a 1-2-3 ternary system, it should be expected that, this ternary will reduce to a 1-2 binary, and the ternary thermodynamic properties will become 1-2 binary thermodynamic properties. As mentioned above, the symmetrical model can not satisfy this basic requirement. However, this new model can. Since in this particular situation, systems "12" and "31" in fact represent the same binary solution "12" and solution "23" becomes a pure component "2", hence, one has $G_{12}^E = G_{31}^E$ and $G_{23}^E = 0$. According to Eqs.(6) and (7), $\eta_I = 0, \eta_{II} \neq 0$, which leads to $\xi_{12} = 0$ and finally, we obtain

$$X_{1(12)} = x_1 + x_3\xi_{12} = x_1 \tag{15}$$

$$X_{2(12)} = 1 - x_1 = x_2 + x_3 \tag{16}$$

On the other hand, Eq.(5) gives

$$\eta_{III} = \int_0^1 (\Delta G_{23}^E - \Delta G_{31}^E)^2 dx_3 = \int_0^1 (\Delta G_{31}^E)^2 dx_3 \neq 0 \tag{17}$$

Substituting it into Eq.(7), we have

$$\xi_{31} = \frac{\eta_{III}}{\eta_{III} + \eta_I} = \frac{\eta_{III}}{\eta_{III}} = 1 \tag{18}$$

and Eq.(10) will be

$$X_{3(31)} = x_3 + x_2\xi_{31} = 1 - x_1 \tag{19}$$

and

$$X_{1(31)} = x_1 \tag{20}$$

Substitution of Eqs.(15), (16) and Eqs.(19),(20) into Eq.(1) and combination with Eq.(2) yield

$$\begin{aligned} \Delta G^E &= W_{12}\Delta G_{12}^E + W_{31}\Delta G_{31}^E + W_{23}\Delta G_{23}^E = W_{12}\Delta G_{12}^E + W_{31}\Delta G_{31}^E \\ &= \frac{x_1 x_2}{X_{1(12)} X_{2(12)}}\Delta G_{12}^E + \frac{x_3 x_1}{X_{1(31)} X_{3(31)}}\Delta G_{31}^E = \frac{x_1(x_2 + x_3)}{x_1(1 - x_1)}\Delta G_{12}^E \\ &= \Delta G_{12}^E \end{aligned} \tag{21}$$

The above result means that, the ternary excess Gibbs energy, under this particular situation, will reduce to a binary excess Gibbs energy as should be expected.

4) In contrast to all asymmetrical models, this new model does not need the human interference in arranging three components to three apexes of a triangle. Unlike an asymmetrical model where the selected binary compositions for three binaries are already fixed before assigning an actual system to it, the selected binary compositions for this new model are always related to the three components themselves. When two components exchange their apex positions in a triangle, the selected binary compositions will also exchange, and as a result, the whole model will be unchanged.

5). This new model has successfully broken down the wall between symmetrical models and asymmetrical models. The example given in the last paragraph "3)", in fact, represents an asymmetric model similar to the Toop model or Hillert model, in which the mole fractions of component "1" in "12" and "13" are the same. Alternatively, if three binaries "12","31","23" have a similar excess Gibbs energy curve, $\xi_{12}, \xi_{31}, \xi_{23}$ may be the same depending on different situations. That will result in a symmetric model. For example, when $\xi_{12} = \xi_{31} = 0.5$, according to Eq.(13), $\xi_{23} = 0.5$, and the new model represents Muggianu's model.

6) In addition to the cases which can be described by symmetrical and asymmetrical models, there are some other cases which meet neither symmetrical conditions nor current asymmetrical conditions. In this particular situation, three binaries have different selected compositions, which still can be described by this new model. In other words, this new model can be used in more practical systems than symmetric plus asymmetric models.

VI. Application to Cu-Mg-Ni Ternary System

So far our discussion is restricted only to a theoretical analysis, and it shows that this new method has advantages almost in every aspect. However, we still need an example to prove that this model is better in practical use.

Recently, Gnanasekaran and Ipser have studied the Cu-Mg-Ni ternary system by using an isopiestic method[19]. They offered a good set of data and tried to use a theoretical model to compare their experimental results. The authors noted that, in this ternary system two out of the three binary systems are characterized by strong affinities, therefore, they decide to use Bonnier model[5] to calculate ternary excess Gibbs energies. Their calculated results show that the agreement between experimental data and theoretical prediction is not satisfactory. In order to eliminate these differences, the authors added a ternary interaction term in their formalism. However, the parameters of this new added interaction term actually are unknown and must be estimated from a least square fitting. In other words, their treatment is still difficult to use because of these unknown parameters.

In this section, the new model presented here will be used to see how well it works. First the three binary data for Cu-Mg, Cu-Ni and Mg-Ni have been used to estimate ξ_{ij} in terms of Eqs.(6) to (8). According to the reference[19], the excess Gibbs energy of mixing for 12, 23 and 31 will be[20-22]

$$\Delta G^{E}_{12}(J/mol) = X_1X_2[(-37871.16+5.72757T)+8415.79(X_1-X_2)] \tag{22}$$

$$\Delta G^{E}_{23}(J/mol) = X_2X_3[(12007.7+1.95556T)+(-2083.6+1.10113T)\cdot(X_2-X_3)+(449.6+0.46893T)(X_2-X_3)^2] \tag{23}$$

$$\Delta G^{E}_{31}(J/mol) = X_3X_1[-60218.199+26.009T] \tag{24}$$

where 1,2 and 3 represent Mg, Cu and Ni respectively. Substituting Eqs.(22) to (24) into Eqs.(3) to (5), one can find η_{I}=406680.2, η_{II}=69415920 and η_{III}=64591018. Combining these η with Eqs.(6) to (8), we obtain $\xi_{12} = 0.005824, \xi_{23} = 0.518002$ and $\xi_{31} = 0.993743$. In order to check the correction of calculated ξ, the Eq.(13) has been used,

$$(1-\xi_{12})(1-\xi_{31})(1-\xi_{23}) = \xi_{12}\xi_{31}\xi_{23} = 0.002998 \tag{25}$$

The above result shows that the calculations of ξ are correct. Then substituting these ξ_{ij} into Eqs.(9) to (11), the selected binary compositions can be obtained. Finally, the ternary excess Gibbs energy of mixing can be calculated through Eq.(12). The calculated results have been plotted in Figure 3. For comparison, the Gnanasekaran and Ipser's data as well as the curve drawn by them using the Bonnier model are also included, from which it may be seen that the line calculated from new model is much closer to the experimental data than that predicted by Bonnier model. Figure 3 is calculated for the condition of x_{Cu} / x_{Ni}=1.0. Similar results have been obtained under the pseudo binary lines x_{Cu} / x_{Ni}=2 and 0.5, which are shown in Figure 3 too. It should be mentioned here that, for the plot of x_{Cu} / x_{Ni}=0.5, there exists an experimental point in the left

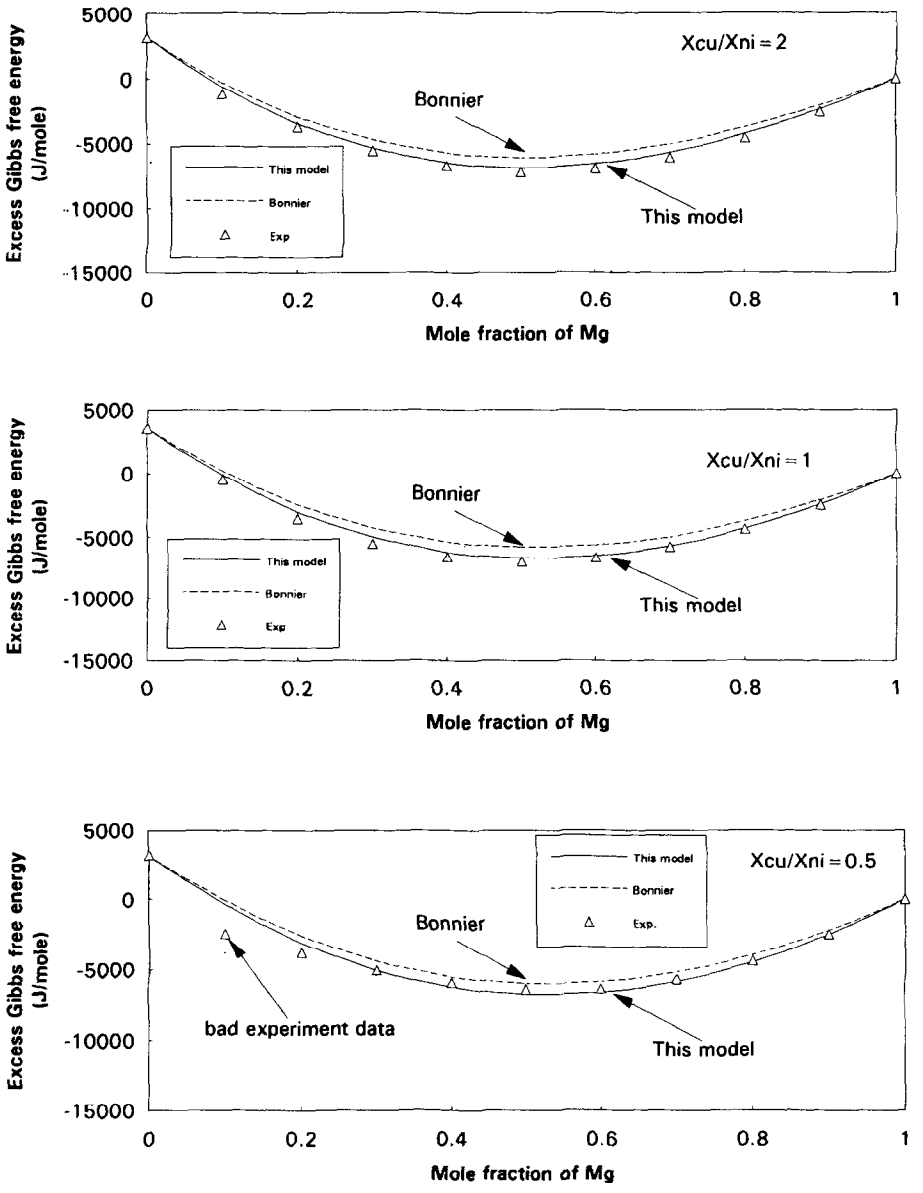


Figure 3. Excess Gibbs energy of mixing at 1173K for liquid Mg-Cu-Ni ternary system

hand side of plot, which is abnormally lower than other points. This is likely to be due to experimental error since if one draws a curve for experimental data, there will be a discontinuity at this point. In summary, it may be seen that, this new model is much better than Bonnier model in the whole concentration range. It is also noted that, since the Bonnier model actually is an asymmetric model, a different arrangement of three components Cu,Mg,Ni in three apexes using that model will cause a different result. In fact, another arrangement probably will get even worse results. However, this new model doesn't require any arrangement for three components. Similar results are also obtain if the Kohler and Muggianu models are applied to the Cu-Mg-Ni system, in which the Gibbs energies predicted by this new model are also closer to experimental data than those estimated from both Kohler and Muggianu models.

As a matter of fact, the Bonnier model is not the best choice for this system, probably the Toop or the Hillert model will be better as analyzed by Hillert[9]. This conclusion now can be obtained directly from this new model, since the three similarity coefficients are approximately equal to zero, unity and 0.5 respectively ($\xi_{12} \approx 0.0058 \approx 0$, $\xi_{31} \approx 0.994 \approx 1$ and $\xi_{23} \approx 0.518 \approx 0.5$), and this new model actually will reduce to the Toop or Hillert models.

VII Conclusions

(1) The model presented in this paper breaks the boundary between symmetric and asymmetric models and simplifies various kinds of models to one.

(2) This new model doesn't require any human interference in selecting models and arranging the three components to apexes of composition triangle. As a result, the application of computers to the calculation of thermodynamic properties and phase diagrams will become more realistic, especially for multicomponent systems with a large number of components. A detailed description about this new model in multicomponent systems will be published in a separate paper[23].

(3) This new model is more reasonable theoretically than other current models. It can reduce to any particular form as any limiting case is met. Especially, it can reduce to a binary system as the second and the third components are identical, not like symmetric models that can not reduce to a binary system even if two components are exactly the same.

(4) This new model not only can give the results that are predicted by symmetric or asymmetric models, but also can give some results that can not be obtained from current models, since the ξ_{ij} can change its value from 0 to 1. In other words, the new model can be suitable for more cases.

(5) The application of this new model to the practical system shows that, this new model is feasible in practical applications.

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