



# Applications of an extended Miedema's model for ternary alloys

P.K. Ray\*, M. Akinc, M.J. Kramer

Ames Laboratory and Department of Materials Science and Engineering, Iowa State University, Ames, IA 50011, USA

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## ABSTRACT

The extension of Miedema's semi-empirical model to ternary systems by means of an energy minimization scheme was implemented to demonstrate a number of physical phenomena associated with select ternary alloys. In order to gain a thermodynamic understanding of glass forming ability of Zr based alloys, a combination of extended Miedema's model and lattice strain energies has been invoked. The extended Miedema approach has also been used to study the phase selection during crystallization for amorphous Zr–Cu–Ni alloys. Also extended Miedema's model was used to illustrate its applicability to study the phase stability of Mo–Nb–Si alloys around  $M_3Si$  composition ( $M = Mo, Nb$ ), by predicting the amount of Nb (30 at.%) required to destabilize the structure.

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## 1. Introduction

During the 1970s and early 1980s, Miedema and his colleagues developed a semi-empirical approach that could predict formation enthalpies of binary alloys [1–7]. The Miedema model is built on the “macroscopic atom” picture. The basic assumption in this case is that the reference can be chosen as atoms embedded in a metal, instead of free atoms. The essence of the model lies in estimating quantitatively the effects of the change in the Wigner–Seitz cell boundary electron density. Dissimilar cells, in contact with each other, would tend to shift their electron densities such as to remove the cell boundary discontinuities in order to form the alloy. Elimination of such discontinuities is expected to require energy; hence the electron density difference  $\Delta n_{ws}$  accounts for a positive contribution to the interface energies. Following the development of Miedema's model, a number of research groups tried to use this model for a variety of metallurgical problems ranging from glass formation to lattice defects [8–19].

Miedema's model, in its original form, is applicable only to binary alloys. Hence, in order to use it for multicomponent systems, a number of different extensions have been proposed [10,11,20,21]. The energy minimization approach which is presented in the following sections involves treating the formation enthalpy as a weighted sum of constituent binary systems, with the weights and the individual binary compositions being chosen such that the net formation enthalpy of the system is minimized. In this paper, we will illustrate application extended Miedema model

to study several physical phenomena associated with ternary alloys.

## 2. Theoretical calculations

In this paper, we adopt an approach similar to that of Gallego, with each of the binaries having a certain weight. These weights are determined by numerical minimization of enthalpy. In this approach, the formation enthalpy is given as:

$$\Delta H = \phi_1 \Delta H_{AB}(c_A) + \phi_2 \Delta H_{BC}(c_B) + \phi_3 \Delta H_{CA}(c_C) \quad (1)$$

where  $c_A$ ,  $c_B$  and  $c_C$  are atom fraction of species  $A$ ,  $B$  and  $C$  in the  $AB$ ,  $BC$  and  $CA$  binaries respectively and  $\phi_1$ ,  $\phi_2$  and  $\phi_3$  are weights assigned to each of the binaries. These weights are found by minimizing  $\Delta H$  under the following set of constraints:

$$\begin{aligned} \sum_{i=1}^3 \phi_i &= 1 \\ \phi_1 c_A + \phi_3 (1 - c_C) &= x_A \\ \phi_2 c_B + \phi_1 (1 - c_A) &= x_B \\ \phi_3 c_C + \phi_2 (1 - c_B) &= x_C \end{aligned} \quad (2)$$

Once the values of the weights and binary compositions have been determined, the formation enthalpy can be easily computed.

The formation enthalpies of the constituent binary systems are calculated using Miedema's semi-empirical model [1–3,5–7,22]. According to this model, the net enthalpy of mixing is given by the relation:

$$\Delta H_{tot} = \Delta H_{ch} + \Delta H_{el} + \Delta H_{st} \quad (3)$$

\* Corresponding author. Tel.: +1 515 294 4064; fax: +1 515 294 5444.  
E-mail address: [prat@iastate.edu](mailto:prat@iastate.edu) (P.K. Ray).

where  $\Delta H_{ch}$ ,  $\Delta H_{el}$ ,  $\Delta H_{st}$  represent the chemical, elastic and structural contributions respectively.

The chemical enthalpy may be evaluated as follows:

$$\Delta H_{ch} = c_A c_B \{ f_B^A \Delta H_{ic}^{AB} + f_A^B \Delta H_{ic}^{BA} \} \quad (4)$$

$f_B^A$  represents the degree to which A is surrounded by B, and is given as:

$$f_B^A = c_B^s [1 + \gamma (c_A^s c_B^s)^2] \quad (5)$$

where  $c_A^s$  represents the concentration of A at the surface and  $\Delta H_{ic}^{AB}$  represents the interfacial enthalpy for A surrounded by B. The factor  $\gamma$  takes the values 8, 5 and 0 for intermetallics, metallic glasses and solid solutions respectively. The surface concentrations may be given as:

$$c_B^s = \frac{c_B V_B^{2/3}}{c_A V_A^{2/3} + c_B V_B^{2/3}} \quad (6)$$

The interfacial enthalpy is given by:

$$\Delta H_{ic}^{AB} = \frac{V_A^{2/3}}{(n_{ws}^{-1/3})_{av}} \{ -P(\Delta\phi^*)^2 + Q(\Delta n_{ws}^{1/3})^2 \} \quad (7)$$

The values of  $P$  and  $Q$  are dependent on the type of metals forming the alloy/intermetallic compound. Usually, the value of  $P$  is taken as 14.2 for metals with valency higher than 2, and 10.7 for metals with valency of 1 or 2. The  $P/Q$  ratio is maintained at 9.4 [1,7]. Eq. (7) has to be modified for alloys of a transition metal with a non-transition metal due to an additional enthalpy term arising out of filling of Brillouin zones of a particular crystal structure. The modified relation may be expressed as:

$$\Delta H_{ic}^{AB} = \frac{V_A^{2/3}}{(n_{ws}^{-1/3})_{av}} \{ -P(\Delta\phi^*)^2 + Q(\Delta n_{ws}^{1/3})^2 - R^* \} \quad (8)$$

The volume changes associated with charge corrections effects may be given as [6]:

$$\Delta V_i = \frac{1.5c_j^s V_i^{2/3} (\phi_i^* - \phi_j^*)}{2(n_{ws}^{-1/3})_{av}} (n_{ws,i}^{-1} - n_{ws,j}^{-1}) \quad (9)$$

$$V_i^* = V_i + \Delta V_i \quad (10)$$

Valence state of element determines the preferred crystal structure that it forms. Hence, during the process of alloying, if the elements are forced to form an alloy having a different crystal structure, an additional structural enthalpy term, needs to be added to the chemical enthalpy term. Miedema et al. proposed that the total structural enthalpy when an element "A" is dissolved in "B" is given by [23]:

$$H_{A \text{ in } B}^{struct} = (Z_A - Z_B) \frac{\partial E^{struct}(B)}{\partial Z} + (E_B^{struct} - E_A^{struct}) \quad (11)$$

Once the structural contribution to the enthalpy is estimated, the formation enthalpy of an A–B binary would be given as:

$$\Delta H(A, B) = x_A x_B [ (f_B^A H_{A \text{ in } B}^{i/c} + f_A^B H_{B \text{ in } A}^{i/c}) + (x_B H_{A \text{ in } B}^{struct} + x_A H_{B \text{ in } A}^{struct}) ] \quad (12)$$

In order to study the lattice strain energies of solid solutions we have used the approach developed by Miracle and Senkov [24,25], and extended to ternary systems by Ray et al. [8,26]. According to this approach, the lattice strain energy of a multicomponent system

can be expressed as:

$$E_{molar}^{el} = \sum_{\substack{j=1, j \neq i \\ p=s, i}}^n \frac{c_j^2}{1 - c_i} E_{ij}^{b \text{ in}} \quad (13)$$

where  $E_{ij}^{b \text{ in}}$  is the lattice strain energy of a binary  $i-j$  solid solution ( $i$  as the matrix species and  $j$  as the solute species).

### 3. Results and discussion

The relations presented in the above section were used to study a number of physical phenomena ranging from glass forming ability in ternary alloys to equilibrium phase assemblage in ternary systems.

#### 3.1. Glass forming ability

Over the years, a number of approaches have been proposed to understand glass formation in metallic systems [8–11,24,27–33]. A number of factors like the  $e/a$  ratio [30,31], lattice strain [8,9,24,26], formation enthalpy [8–11,26] and local geometry [8,27–29] have been proposed to explain the phenomena. In this paper, we have focused on the thermodynamics of glass formation. Assuming the frustration of nucleation of intermetallic crystalline phase, the competing phase for the metallic glass is the solid solution. The driving force for crystallization can therefore be given as:

$$\Delta H^{cryst} = (H_{mg}^{ch} + H_{mg}^{str}) - (H_{ss}^{ch} + H_{ss}^{str} + H_{ss}^{lse}) \quad (14)$$

where  $H_{ss}^{ch}$  is the chemical contribution to enthalpy of a solid solution,  $H_{ss}^{str}$  is the structural contribution to enthalpy of the solid solution,  $H_{ss}^{lse}$  is the lattice strain energy of the solid solution,  $H_{mg}^{ch}$  is the chemical contribution to the enthalpy of a metallic glass and  $H_{mg}^{str}$  is the structural contribution to the enthalpy of a metallic glass. Since glasses are thermodynamically treated as liquids, it is assumed that they have zero lattice strain energy. The higher the value of this driving force for crystallization, the easier it is for the glass to form.

Several experimental approaches have been postulated for measuring glass forming ability (GFA). Ideally, lower the critical cooling rate with which the liquid can still transform to glass, the better is the glass forming ability. With this in mind, a number of parameters involving the liquidus temperature ( $T_l$ ), the crystallization temperature ( $T_x$ ) and the glass transition temperature ( $T_g$ ) have been formulated to assess the GFA of an alloy. The first parameter for assessing GFA was formulated by David Turnbull who used the reduced glass transition temperature ( $T_{rg}$ ), the ratio of  $T_g$  and  $T_l$ , as a measure of GFA [34]. Subsequently, it has been debated [35–37] whether a ratio based on  $T_g/T_l$  is a better measure or  $T_g/T_m$  is a better measure where  $T_m$  is the onset of the melting. In 1978, Donald and Davies suggested [38] the use of  $T_l^{mix}$  as an alternative for  $T_l$  in the expression for  $T_{rg}$ . Here  $T_l^{mix}$  represents the weighted average of the melting points of the individual components. This results in a simple parameter given as:

$$\Delta T^* = \frac{T_l^{mix} - T_m}{T_l^{mix}}, \quad T_l^{mix} = \sum_{i=1}^n x_i T_m^i \quad (15)$$

Recently, Inoue et al. [39] have proposed a new parameter for GFA. It was suggested that  $\Delta T_x$  be used as a parameter for GFA. Better glass formers are expected to exhibit a wider supercooled range or a higher value of  $\Delta T_x$ . Lu and Liu [40] suggested the use of

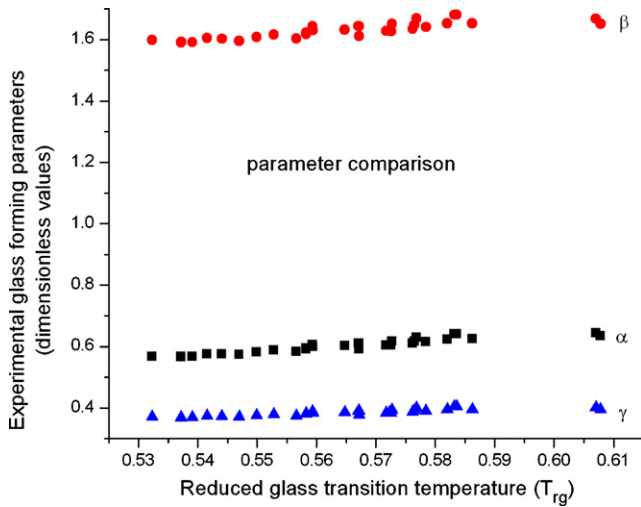


Fig. 1. Comparison of different glass forming parameters.

$\gamma$  parameter, defined as:

$$\gamma = \frac{T_x}{T_g + T_l} \quad (16)$$

As a better glass forming criterion. It is widely accepted that the critical cooling rate ( $R_c$ ) is a good measure of GFA. On this basis, the  $\gamma$  parameter shows lesser scatter as opposed to the parameters suggested by any of the earlier researchers. Mondal and Murty [41] have suggested an even more simplified and effective approach towards GFA. They suggested two parameters:

$$\alpha = \left( \frac{T_x}{T_l} \right) \quad (17)$$

and

$$\beta = \frac{T_x}{T_g} + \frac{T_g}{T_l} \quad (18)$$

using these parameters GFA can be assessed effectively. The former parameter, in fact, does not even require the knowledge of  $T_g$  for predicting GFA. Fig. 1 shows a comparison of  $\alpha$ ,  $\beta$  and  $\gamma$  parameters with respect to the reduced glass transition temperature. It can be seen that the trend is consistent for all of them. Fig. 2a and b shows plots of the driving force for crystallization vis-à-vis the  $\alpha$  and the  $\gamma$  parameters respectively. The  $\gamma$  parameter has been estimated using the experimental data available on Zr based metallic glasses available in the literature [42–44]. The figure is consistent with our expectation that a more negative driving force will aid glass forming ability. Hence, given a glass-forming system, the extended Miedema approach can be used to identify good glass forming composition regimes.

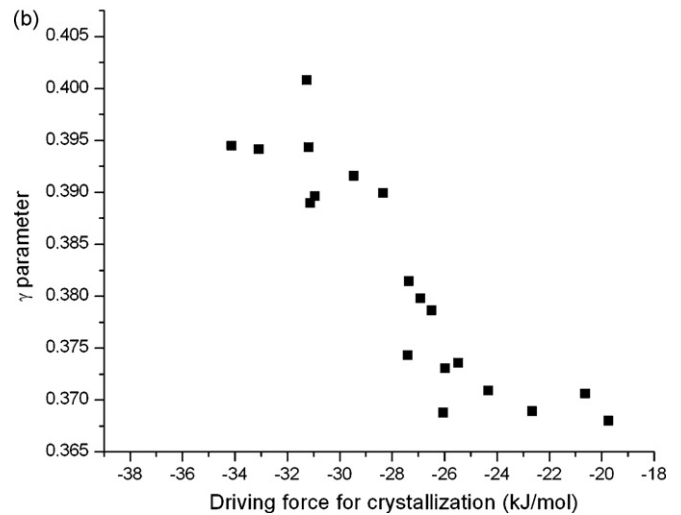
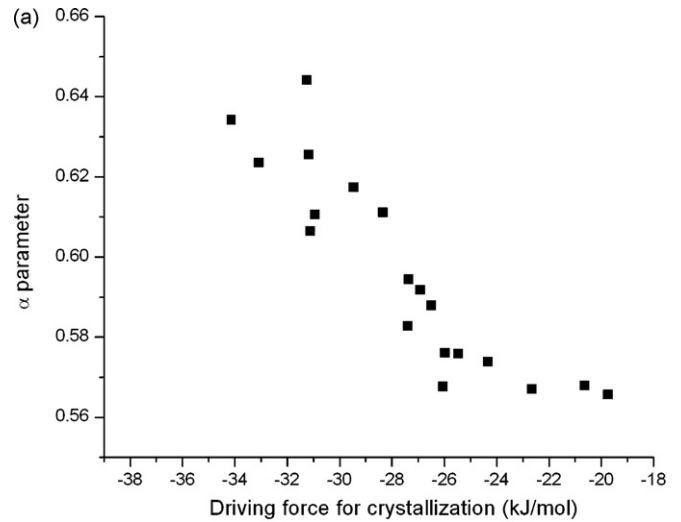


Fig. 2. (a) Comparison of the driving force for crystallization with  $\alpha$  parameter. (b) Comparison of the driving force for crystallization with  $\gamma$  parameter.

### 3.2. Crystallization pathways in metallic glasses

Experimental studies on crystallization behavior of  $Zr_2Cu_{1-x}Ni_x$  metallic glasses have shown that the  $Zr_2Ni$  (c16) based phase crystallizes for  $x \leq 0.625$  [45,46]. In the range  $0.625 \leq x \leq 0.375$ , both  $Zr_2Cu$  and  $Zr_2Ni$  based phases can be observed, while for  $x \leq 0.375$   $Zr_2Cu$  (c11b) is the preferred phase. Table 1 shows a list of the optimized parameters ( $c_A$ ,  $c_B$ ,  $\varphi_1$  and  $\varphi_2$ ). The Cu–Ni binary has a negligible contribution to the formation enthalpy, and the fraction contribution of this binary ( $\varphi_3$ ) is 0.01. The major contributions to the formation enthalpy comes from the Zr–Cu binary for values of  $x$  lower than 0.375, with the binary composition being approximately

Table 1  
Fraction of Zr–Ni and Zr–Cu binary in the Zr–Cu–Ni ternary and their respective compositions.

| Composition            | Fraction cluster A (Zr–Ni) $\varphi_1$ | Cluster composition (atom fraction Zr) $c_A$ | Fraction cluster B (Zr–Cu) $\varphi_2$ | Cluster composition (atom fraction Cu) $c_B$ |
|------------------------|--|--|--|--|
| $Zr_2Ni_{1/8}Cu_{7/8}$ | 0.11                                   | 0.7  | 0.88                                   | 0.33   |
| $Zr_2Ni_{1/4}Cu_{3/4}$ | 0.19                                   | 0.6  | 0.80                                   | 0.31   |
| $Zr_2Ni_{3/8}Cu_{5/8}$ | 0.29                                   | 0.6  | 0.70                                   | 0.30   |
| $Zr_2Ni_{1/2}Cu_{1/2}$ | 0.40                                   | 0.6  | 0.59                                   | 0.28   |
| $Zr_2Ni_{5/8}Cu_{3/8}$ | 0.50                                   | 0.6  | 0.49                                   | 0.25   |
| $Zr_2Ni_{3/4}Cu_{1/4}$ | 0.60                                   | 0.6  | 0.39                                   | 0.21   |
| $Zr_2Ni_{7/8}Cu_{1/8}$ | 0.71                                   | 0.6  | 0.28                                   | 0.14   |

Zr<sub>2</sub>Cu, where as the major contribution for values of  $x$  higher than 0.625 is due to the Zr–Ni binary.

Cu and Ni have a difference of one valence electron with a minor difference in size. According to Miedema's model, the interaction parameter between different species is governed by volume differences and differences in electron density at the Wigner–Seitz cell boundary. The volume difference being negligible, the difference in the number of valence electron and consequently, a difference in the Wigner–Seitz cell boundary electron density plays a key role in Zr–Cu and Zr–Ni systems having different interaction parameters. Since the formation enthalpy of alloys in the Cu–Ni binary is fairly small, the formation enthalpy of the ternary glass is mainly decided by contributions of the Zr–Cu and Zr–Ni binaries. A more negative Zr–Ni interaction parameter should result in the enthalpy minimization routine favoring a relatively greater phase width for the c16 (Zr<sub>2</sub>Ni) structure as compared to the c11b (Zr<sub>2</sub>Cu) structure. This has already been verified experimentally by Kramer et al. [45]. This indicates that at least in this system, the electronic structure plays an important role in GFA, a fact ignored in other parameterization schemes.

### 3.3. Phase stability in ternary alloy systems

Mo–Si based alloys constitute an important class of ultra-high temperature alloys [47–49]. However, a key impediment to widespread usage of these materials is the relatively low fracture toughness. Microstructural investigations show the presence of a three-phase alloy, with the three phases being a Mo based solid solution, a M<sub>3</sub>Si (M = metal) phase and M<sub>5</sub>Si<sub>3</sub> phase [50–52]. It has been shown that the M<sub>3</sub>Si phase has poor mechanical properties [53,54]. It was surmised that the removal of this phase could lead to improved toughness. Using the present model, formation enthalpies of the M<sub>3</sub>Si phase and a phase mixture of Mo based solid solution and M<sub>5</sub>Si<sub>3</sub> was calculated. Assuming limiting conditions, when M<sub>3</sub>Si exists in equilibrium with Mo and M<sub>5</sub>Si<sub>3</sub> [55]:



Here,  $x_a$  and  $x_b$  represent the atom percentage of Mo and Nb,  $\lambda$  and  $\psi$  denote the phase fraction of the solid solution and M<sub>5</sub>Si<sub>3</sub>,  $\varphi_1$  and  $\varphi_2$  represent the atom percentage of Mo and Nb in the solid solution and  $\mu_1$  and  $\mu_2$  represent the atom percentage of Mo and Nb in the M<sub>5</sub>Si<sub>3</sub> phase. Using principles of mass balance, we have the following set of equations:

$$x_a + x_b = 75 \quad (19a)$$

$$\lambda\varphi_1 + \psi\mu_1 = x_a \quad (19b)$$

$$\lambda\varphi_2 + \psi\mu_2 = x_b \quad (19c)$$

$$\lambda + 37.5\psi = 25 \quad (19d)$$

$$\varphi_1 + \varphi_2 = 99 \quad (19e)$$

$$\mu_1 + \mu_2 = 62.5 \quad (19f)$$

Solving the equations, we obtain  $\psi = 2400/3650$  and  $\lambda = 1250/3650$ . The remaining variables can be expressed in terms of  $\varphi_1$ .

$$\mu_1 = \frac{x_a - \lambda\varphi_1}{\psi} \quad (20a)$$

$$\varphi_2 = 99 - \varphi_1 \quad (20b)$$

$$\mu_2 = 62.5 - \frac{x_a - \lambda\varphi_1}{\psi} \quad (20c)$$

Using these relations, the enthalpy difference between the A15 phase and an assemblage of solid solution and M<sub>5</sub>Si<sub>3</sub> phases and

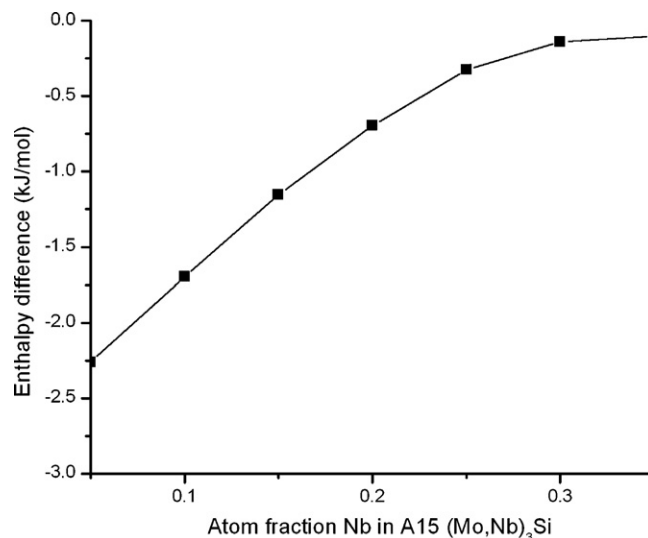


Fig. 3. Plot of enthalpy difference between M<sub>3</sub>Si phase and a phase mixture of Mo based solid solution and M<sub>5</sub>Si<sub>3</sub> as a function of Nb addition. The shaded region indicates the composition range where the M<sub>3</sub>Si phase gets destabilized.

plotted in Fig. 3. From this figure it can be seen that the difference in formation enthalpy of the (Mo,Nb)<sub>3</sub>Si phase and the phase assemblage of (Mo,Nb,Si) solid solution + (Mo,Nb)<sub>5</sub>Si<sub>3</sub> decreases with increasing Nb addition and tends to zero at approximately 30 at.% Nb. Experimental studies have indicated that the (Mo,Nb)<sub>3</sub>Si phase does indeed get destabilized when further Nb is added [55].

## 4. Conclusions

In this paper we have successfully applied the extension of Miedema's semi-empirical model in our previous work to a number of physical phenomena. We were able to understand glass forming ability of a number of alloy systems using thermodynamic parameters based on the formation enthalpies estimated using the extended Miedema model. Based on the individual binaries, we could obtain trends in the predominant clusters present in glasses and thus gain an understanding of the crystallization pathways. The model could also be used to understand phase stability issues in the refractory metal silicides. Thus it can be seen that the model is capable of giving good approximate predictions and has the potential to be applied in diverse areas.

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