



Theoretical prediction of bulk glass forming ability (BGFA) of Ti–Cu based multicomponent alloys

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ABSTRACT

The bulk glass forming ability (BGFA) of Ti–Cu based multicomponent alloys has been evaluated via theoretical modeling and computer simulation studies based on a combination of electronic theory of alloys in the pseudopotential approximation and the statistical thermodynamical theory of liquid alloys. The magnitude of atomic ordering energies, calculated by means of the electronic theory of alloys in the pseudopotential approximation, was subsequently used for calculation of the key thermodynamic parameters such as enthalpy, entropy and Gibbs free energy of mixing, viscosity, and critical cooling rate of the binary Ti–Cu and ternary Ti–Cu–X alloys. The potential alloying elements (X) can be divided in two groups defined by their effect on the variation of the negative heat of mixing and their influence on the critical cooling rate. Most of the predicted candidate alloying elements from either X_I (Al, Si, Ag) or X_{II} (Co, Ni, Fe, Sn, Be) and/or both groups have already been used successfully for the fabrication of new Ti–Cu based bulk metallic glasses. It was also shown that the critical cooling rate appears to be a more important parameter rather than the change in the negative heat of mixing for the prediction of candidate alloying elements improving BGFA.

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1. Introduction

Bulk metallic glasses (BMGs) are the main research interests of scientists for the last 10–15 years because of their unique material properties and characteristics in comparison with their crystalline counterparts. Since the discovery of a series of glassy alloys with large glass forming ability in Ln–Al–TM, Mg–Ln–TM, Zr–Al–TM, Ti–Zr–TM, Hf–Al–TM, Ti–Zr–Al–Be–TM, and Ti–Zr–Be–TM (Ln = lanthanide metals, TM = transition metals) systems, bulk glassy alloys have attracted rapidly increasing interest because of their importance to both materials science and engineering application [1–11]. BMGs have a wide range of interesting properties, including near theoretical strength, high hardness, extremely low damping characteristics, excellent wear properties, high corrosion resistance and low shrinkage during cooling. Among the above-described alloy systems, copper based bulk metallic glasses have become especially attractive in the BMG class due to their relatively low cost and improved mechanical properties [12–22]. These alloys display enhanced glass forming ability; accordingly they lead to the development of commercial bulk metallic glasses for several applications [9–15]. Recently, titanium based bulk amorphous alloys also began to attract the attention of the scientists

and engineers due to their high specific strength, good corrosion resistance and high-temperature properties [23–40].

A number of semi-empirical criteria and theoretical approaches have been proposed to evaluate bulk glass forming ability (BGFA) of the multicomponent alloy systems [1,41–44]. In spite of great efforts that have been concentrated on the analysis of bulk glass forming ability (BGFA) of various multicomponent alloy systems, the exact nature of the mechanism of BGFA has not been revealed and is largely empirical. Hence understanding the mechanism behind the easy glass formation and/or high glass forming ability in metallic alloy systems is crucial for the development of new BMGs. In this study, therefore, we present what we believe to be the first attempt to assess the BGFA of Ti–Cu based alloys the atomic level by combining the electronic theory of alloys in the pseudopotential approximation with statistical thermodynamical theory of liquid alloys.

2. Theory

The magnitude and sign of atomic ordering energies of atomic pairs in the binary and multicomponent alloy systems directly influences the key thermodynamic parameters. An increase in the ordering energy of an alloy system will increase both the enthalpy and entropy of mixing and viscosity, but will reduce the critical cooling rate necessary to prevent nucleation and growth of competing crystalline phases to preserve the amorphous

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structure of the melt during solidification [45,46]. Therefore, determination of the atomic ordering energy is essential and can be employed for the estimation of BGFA of alloy systems in the search of new BMGs.

The partial ordering energies $W_{\alpha\alpha'}(R)$ based on electronic theory of multicomponent alloys in the pseudopotential approximation can be calculated using the following equations [47–50]

$$W_{\alpha\alpha'}(R_1) = \frac{\bar{\Omega}_0}{\pi^2} \int_0^\infty F_{\alpha\alpha'}(q) \frac{\text{Sin}(qR_1)}{qR_1} q^2 dq, \quad (1)$$

where

$$F_{\alpha\alpha'}(q) = -\frac{\bar{\Omega}_0}{8\pi} |\omega_\alpha^0(q) - \omega_{\alpha'}^0(q)|^2 q^2 \frac{\varepsilon(q) - 1}{\varepsilon^*(q)} + \frac{2\pi}{\bar{\Omega}_0 q^2} |Z_\alpha^* - Z_{\alpha'}^*|^2 \exp\left(-\frac{q^2}{4\zeta}\right). \quad (2)$$

In Eqs. (1) and (2) $\bar{\Omega}_0$ is the average atomic volume of the ternary alloy; $\varepsilon(q)$ is the dielectric constant in the Hartree approximation; $\varepsilon^*(q)$ is the modified dielectric constant which takes into account the correlation and exchange effects; $\omega_\alpha^0(q)$ and $\omega_{\alpha'}^0(q)$ are the form-factors of unscreened pseudopotentials of α and α' component ions, respectively; Z_α^* ($Z_{\alpha'}^*$) is the effective valency of the α (α') component atoms and ζ is the Ewald parameter.

Free energy of mixing, ΔG^M in terms of enthalpy of mixing, ΔH^M and entropy of mixing ΔS^M is defined as [51]:

$$\Delta G^M = \Delta H^M - T\Delta S^M. \quad (3)$$

Entropy of mixing, ΔS^M is related to the ideal configurational entropy, ΔS^{ideal} and mismatch term of entropy, S_σ [51]:

$$\Delta S^M = \Delta S^{\text{ideal}} + S_\sigma. \quad (4)$$

Using the regular solution model, ΔH^M for the multicomponent systems with N elements can be calculated through following equation [51]:

$$\Delta H^M = \sum_{i=1, i \neq j}^N \gamma_{ij} C_i C_j, \quad (5)$$

where γ_{ij} is the regular solution interaction parameter between i and j elements, and C_i and C_j are the atomic fractions of i and j elements, respectively. Regular solution interaction parameter is assumed to be both temperature and composition independent however; it is related to coordination number (Z) and ordering energy (W) via:

$$\gamma_{ij} = \frac{ZW}{2}. \quad (6)$$

At the same time, according to the regular solution theory, ΔS^{ideal} for the multicomponent systems with N elements was determined as [51]:

$$\Delta S^{\text{ideal}} = -R \sum_{i=1}^N (C_i \ln C_i), \quad (7)$$

where R is the gas constant.

In order to calculate entropy of mixing, mismatch term of entropy should also be determined. An equation of state for the mixture of hard spheres was proposed by Mansoori et al. [45] containing ideal gas-, concentration-, packing- and misfit-terms. However, in the present study, only the mismatch term of entropy was taken into account by using following equation [52]:

$$S_\sigma = k_B \left\{ \frac{3}{2} (\zeta^2 - 1) y_1 + \frac{3}{2} (\zeta - 1)^2 y_2 - \left[\frac{1}{2} (\zeta - 1) (\zeta - 3) + \ln \zeta \right] (1 - y_3) \right\}, \quad (8)$$

Where $\zeta = (1 - v)^{-1}$ and v is the packing fraction. Packing fraction v was taken to be equal to 0.64 assuming random close packed structure [53,54]. Following relations were derived for the multicomponent systems with N elements [51]:

$$y_1 = \frac{1}{\sigma^3} \sum_{j>i=1}^N (d_i + d_j)(d_i - d_j)^2 C_i C_j, \quad (9)$$

$$y_2 = \frac{\sigma^2}{(\sigma^3)^2} \sum_{j>i=1}^N d_i d_j (d_i - d_j)^2 C_i C_j, \quad (10)$$

$$y_3 = \frac{(\sigma^2)^3}{(\sigma^3)^3}, \quad (11)$$

$$\sigma^k = \sum_{i=1}^N C_i d_i^k, \quad (12)$$

where d_i is the atomic diameter of the i th element and $k = 2, 3$.

Calculations of these key thermodynamic parameters enable one to predict the critical cooling rate, which plays an important role in the evaluation of the BGFA. In the present study, for the calculation of critical cooling rate following equation was used [51]:

$$R_c = \psi \frac{k_B T_m^2}{a^3 \eta_{T=T_m}} \exp \left[f_1 \left(\frac{\Delta H^M - T_m \Delta S^{\text{ideal}}}{300R} \right) - f_2 \left(\frac{T_m S_\sigma}{300R} \right) \right], \quad (13)$$

where ψ is a constant (2×10^{-6}), k_B is the Boltzmann constant, T_m is the melting temperature, $\eta_{T=T_m}$ is the viscosity at the melting temperature, a is the interatomic distance and f_1 and f_2 are the fitting parameters. Fitting parameters of f_1 and f_2 were calculated as 0.75 and 1.2, respectively, by using the method of least-squares for 300 K [51]. Viscosity at the melting temperature was calculated by using equation proposed by Battezzati and Greer such as [55]:

$$\eta_{T_m} = C_A \frac{\sqrt{AT_m}}{V^{2/3}}, \quad (14)$$

C_A is a constant of 1.85×10^{-7} (J/K mol^{1/3})^{1/2}, A is the atomic weight, and V is the molar volume at T_m .

Viscosity of liquid metals and alloys is also important in understanding the glass formation of the alloy systems. During undercooling, a rapid increase of viscosity promotes the glass forming ability, and such a phenomenon may be necessary in some composition ranges to express the variation of glass formability in alloys [55]. The change in viscosity of liquid metal can be represented as [56,57]:

$$\frac{\Delta \eta}{\eta} = -\frac{\Delta H^M}{RT}. \quad (15)$$

It should be noted that change in viscosity is positive for ordered alloys since ΔH^M is always negative for glass forming alloy systems.

3. Results

In this present study Ti₆₅Cu₃₅ binary alloy was chosen as the model system to predict the effect of various ternary alloying additions (X) on the BGFA of the Ti₆₄Cu₃₅X₁ alloys. Ordering energy for binary Ti₆₅Cu₃₅ model alloy and partial ordering energies between constituent element atoms of Ti₆₄Cu₃₅X₁ ternary alloys have been calculated for large number of X alloying elements in the periodic table. The calculated partial ordering energies, based on Eqs. (1) and (2), of $W_{\text{Ti-Cu}}(R)$, $W_{\text{Ti-X}}(R)$ and $W_{\text{Cu-X}}(R)$ as a function of interatomic distance in Ti₆₄Cu₃₅X₁ alloy are given in Fig. 1 for X = Ag, Au, Al, and Mn as an example. The nearest neighbor distance between

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