

Optimization of bulk metallic glass forming compositions in Zr–Cu–Al system by thermodynamic modeling

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Abstract

Compositions with high glass forming ability have been identified in Zr–Cu–Al system with the help of a thermodynamic parameter, which takes into account the enthalpy of chemical mixing (ΔH^{chem}), the mismatch entropy normalized by Boltzmann's constant (S_{σ}/k_B) and the configurational entropy (S_{config}/R). The best bulk metallic glass forming composition is identified as the one at ΔH^{chem} and S_{σ}/k_B maxima in a specific range of S_{config}/R . A product of thermodynamic parameters ($\Delta H^{\text{chem}} \times S_{\sigma}/k_B$) is found to have strong correlation with glass forming ability and can help to identify the exact composition that can form bulk metallic glass.

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1. Introduction

Since the discovery of metallic glass [1] and bulk metallic glasses (BMG) [2–4], thermodynamics have been an important tool to understand the science of glass formation [5–9]. A number of glass forming criteria have been developed based on the different phase transition temperatures such as, T_g (glass transition temperature), T_x (onset crystallization temperature), T_l (onset liquidus temperature) and T_m (melting temperature), which are difficult to calculate theoretically [5–9]. Glassy phase at room temperature is also said to have long-range disorder, which is mainly contributed by mismatch [10] and configurational [11] entropies, which cannot be ignored. In earlier thermodynamic approaches [10,12], theoretical glass-forming ranges for ternary alloys were calculated using Miedema's model, which were compared to those obtained by experiments. In the present approach, a thermodynamic parameter has been developed to identify excellent BMG composition using enthalpy of chemical mixing (ΔH^{chem}), the mismatch

entropy normalized by Boltzmann's constant (S_{σ}/k_B) and the configurational entropy (S_{config}/R) and it has been demonstrated for the Zr–Cu–Al based ternary system. Earlier studies have used minor alloying technique [13], microstructure based approach [14] and cluster model [15] to identify the best BMG composition in similar systems. The present approach uses the isometric contours of ΔH^{chem} , S_{σ}/k_B and S_{config}/R and can predict the best BMG composition more closely than the earlier models.

2. Calculation method for thermodynamic parameters

2.1. Enthalpy of chemical mixing

Enthalpy of chemical mixing (ΔH^{chem}) was calculated based on extended regular solution model for ternary systems by equiatomic (Takeuchi's approach) [10] and Gallego's approach [16].

According to equiatomic approach:

$$\Delta H^{\text{chem}} = \sum_{\substack{i=1 \\ i \neq j}}^3 \Omega_{ij} C_i C_j \quad (1)$$

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where Ω_{ij} is the regular solution interaction parameter between i th and j th elements and can be correlated to ΔH^{mix} as

$$\Delta H_{\text{AB}}^{\text{mix}} = X_{\text{A}}X_{\text{B}}\Omega_{\text{AB}} \quad (2)$$

Considering the equiatomic composition in binary A–B system $X_{\text{A}}X_{\text{B}} = 1/4$.

According to Gallego’s approach enthalpy due to chemical contribution can be calculated by

$$\Delta H_{\text{ABC}}^{\text{chem}} = \Delta H_{\text{AB}}^{\text{c}} + \Delta H_{\text{BC}}^{\text{c}} + \Delta H_{\text{AC}}^{\text{c}} \quad (3)$$

where

$$\Delta H_{ij}^{\text{c}} = x_i x_j \left(x_j \Delta H_{ijn}^{\text{mix}} + x_i \Delta H_{jmi}^{\text{mix}} \right) \quad (4)$$

The value of enthalpy due to mixing ΔH^{mix} is calculated from Miedema’s semi-empirical model [17] and substituted for Ω_{ij} . In the Miedema’s model, the mixing enthalpy for infinite dilute solution of two metals A and B, which is also regarded as interfacial enthalpy of atomic cells is given by Eq. (5) and is tabulated elsewhere [17]:

$$\Delta H_{\text{A}}^{\infty} (\text{in B}) = \Delta H^{\text{inter}} (\text{A in B}) = \frac{2V_{\text{A}}^{2/3}}{\left\{ (n_{\text{ws}}^{\text{A}})^{-1/3} + (n_{\text{ws}}^{\text{B}})^{-1/3} \right\}} \times \left[-P(\Delta\phi^*)^2 + Q(\Delta n_{\text{ws}}^{1/3})^2 - R^* \right] \quad (5)$$

where V_{A} is the molar volume of atom A, n_{ws}^{A} and n_{ws}^{B} , the electron density of atomic cells, P and Q are constants, R^* is a correction value for an alloy of a transition metal with one of the polyvalent non-transition metals and $\Delta\phi^*$ is the difference of the modified electronegativities between dissimilar atomic cells of A and B atoms.

2.2. Mismatch entropy

The effect of atomic size mismatch in a system was calculated by empirical relation given by Mansoori et al. [18]. According to this formalism the mismatch entropy was calculated by misfit term S_{σ} from the solution of the Perkus-Yevik integral equation:

$$S_{\sigma} = k_{\text{B}} \left[\frac{3}{2}(\zeta^2 - 1)y_1 + \frac{3}{2}(\zeta - 1)^2 y_2 - \left\{ \frac{1}{2}(\zeta - 1)(\zeta - 3) + \ln \zeta \right\} (1 - y_3) \right] \quad (6)$$

where k_{B} is the Boltzmann’s constant and parameter ζ is defined as $\zeta = 1/(1 - \xi)$ and ξ is packing fraction. In the present study ξ is taken as 0.64, which implies a dense random packing. Dimensionless parameter y_1 , y_2 and y_3 are calculated according to following equation:

$$y_1 = \frac{1}{\sigma^3} \sum_{j \geq i=1}^n (d_i + d_j)(d_i - d_j)^2 c_i c_j \quad (7)$$

$$y_2 = \frac{\sigma^2}{(\sigma^3)^2} \sum_{j \geq i=1}^n (d_i d_j)(d_i - d_j)^2 c_i c_j \quad (8)$$

$$y_3 = \frac{(\sigma^2)^3}{(\sigma^3)^2} \quad (9)$$

$$\sigma^k = \sum_{i=1}^n c_i^k d_i^k \quad (k = 2, 3) \quad (10)$$

Here d_i and d_j are the atomic diameters of i th and j th elements can be calculated from standard data [19]. Configurational entropy is given by

$$S_{\text{config}} = -R \sum_{i=1}^n x_i \ln x_i \quad (11)$$

where x_i is the atomic percent of the i th element.

3. Experimental details

Ingots of Zr–Cu–Al alloys, as tabulated in Table 1, were prepared by arc melting the mixtures of constituent elements under argon atmosphere. The purities of elements are 99.99 wt.% for Cu, 99.9 wt.% for Zr and 99.999 wt.% for Al. Alloy rods with diameter of 3 mm were prepared by means of copper mould suction casting. Structural characterization of these alloys was done by X-ray diffractometry (XRD) using Cu $K\alpha$ radiation ($\lambda = 0.15406$ nm).

4. Results and discussion

From the experiments carried out by various groups (Table 2) it is observed that the S_{config}/R for ternary BMGs containing metals ranges between 0.8 and 1.0 and systems with metalloids like B, C, Ge, Si and P ranges from 0.6 to 1.0. It is well known that the metallic glasses are configurationally frozen supercooled liquids and they are more prone to form at composition near to deep eutectic. It was observed from the previous study [14] that the glass forming ability in

Table 1
 $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ values and phase analysis of the alloys of the present study

S. no.	Alloy composition	$\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ (kJ/mol)	Phase present
1	Zr ₆₆ Cu _{8.5} Al _{25.5}	–3.5	Crystalline
2	Zr ₅₇ Cu _{6.5} Al _{36.5}	–4.0	Crystalline
3	Zr _{40.7} Cu _{7.5} Al _{51.5}	–4.0	Crystalline
4	Zr ₃₁ Cu ₁₀ Al ₆₀	–3.5	Crystalline
5	Zr ₂₄ Cu ₁₃ Al ₆₃	–3.0	Crystalline
6	Zr ₆₅ Cu ₁₉ Al ₁₆	–4.5	Crystalline
7	Zr ₆₁ Cu ₃₂ Al ₇	–5.0	Amorphous
8	Zr _{54.5} Cu ₄₂ Al _{3.5}	–5.7	Amorphous
9	Zr ₄₅ Cu ₄₉ Al ₆	–6.7	Amorphous
10	Zr ₃₆ Cu ₅₈ Al ₆	–5.7	Amorphous

Table 2
 S_{config}/R values for various experimental BMG compositions

Alloy compositions	R_c (K/s)	Z_c (mm)	S_{config}/R	Ref.
Mg ₇₅ Ni ₁₅ Nd ₁₀	46.1	2.8	0.7	[20]
Mg ₇₀ Ni ₁₅ Nd ₁₅	178.2	1.5	0.8	[20]
Mg ₆₅ Ni ₂₀ Nd ₁₅	30.0	3.5	0.9	[20]
Mg ₆₅ Ni ₂₁ Pr ₁₄	—	3.0	0.9	[21]
Mg ₆₅ Cu ₂₅ Y ₁₀	50.0	7.0	0.8	[22]
La ₅₅ Al ₂₅ Ni ₂₀	67.5	3.0	0.9	[23]
La ₅₅ Al ₂₅ Cu ₂₀	72.3	3.0	0.9	[23]
La ₆₆ Al ₁₄ Cu ₂₀	37.5	2.0	0.8	[24]
Pd ₇₇ Cu ₆ Si ₁₇	125.0	2.0	0.7	[2]
Pd _{77.5} Cu ₆ Si _{16.5}	100.0	1.5	0.6	[25]
Pd _{73.5} Cu ₁₀ Si _{16.5}	—	2.0	0.7	[2]
Pd _{71.5} Cu ₁₂ Si _{16.5}	—	2.0	0.8	[2]
Pd ₄₀ Ni ₄₀ P ₂₀	0.16	25.0	1.0	[26]
Cu ₆₀ Zr ₃₀ Ti ₁₀	—	4.0	0.9	[27]
Cu ₆₀ Zr ₂₂ Ti ₁₈	—	3.0	0.9	[28]
Cu ₄₈ Zr ₄₈ Al ₄	—	5.0	0.8	[29]
Zr ₄₇ Cu ₄₇ Al ₆	—	5.0	0.9	[29]
Zr ₄₅ Cu ₄₅ Al ₁₀	—	3.0	0.9	[29]
Zr ₄₆ Cu ₄₆ Al ₈	—	>5.0	0.9	[29]
Zr ₆₀ Ni ₁₉ Al ₂₁	—	3.0	0.9	[30]
Zr ₅₃ Co _{23.5} Al _{23.5}	—	3.0	1.0	[31]

Zr–Cu–Al system falls in a broad range from Zr rich to Cu rich end. In spite of this, interestingly, S_{config}/R value for all the BMG compositions lies between a narrow range of 0.8–1.0. For the compositions where metalloids are present, not only there is decrease in eutectic temperature in general but also an increase in BMG forming ability, which is reflected in their wider range of S_{config}/R as shown in Table 2. In the present study, it is assumed that S_{config}/R should be in the range of 0.8–1.0 for high glass forming ability of metal–metal systems as is observed in Table 1. Fig. 1 shows the isometric S_{config}/R contours for an A–B–C system. It can be seen from Eq. (11) that S_{config}/R depends on the composition and does not depend on the alloy system.

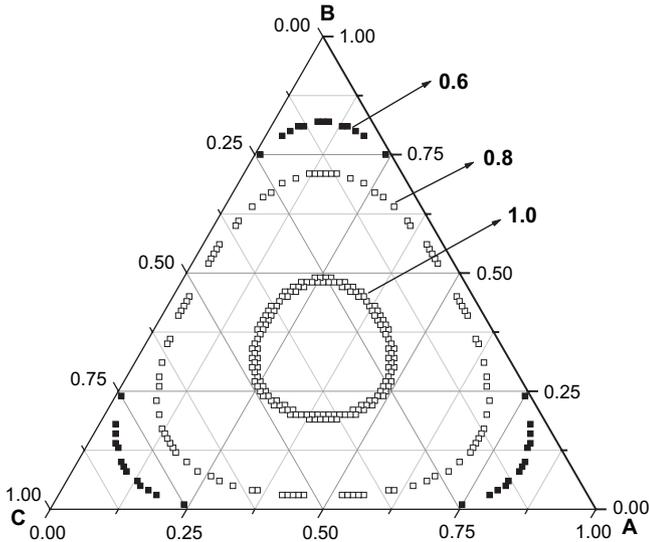


Fig. 1. Isometric contours of configurational entropy, S_{config}/R .

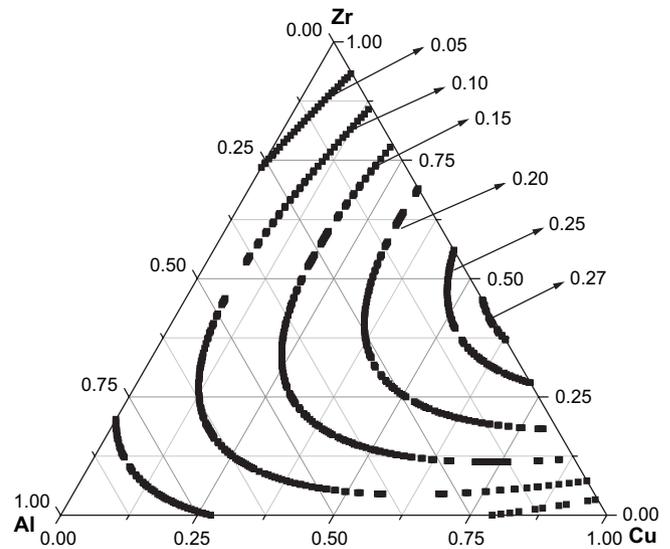


Fig. 2. Isometric contours of mismatch entropy normalized by Boltzmann's constant, S_o/k_B .

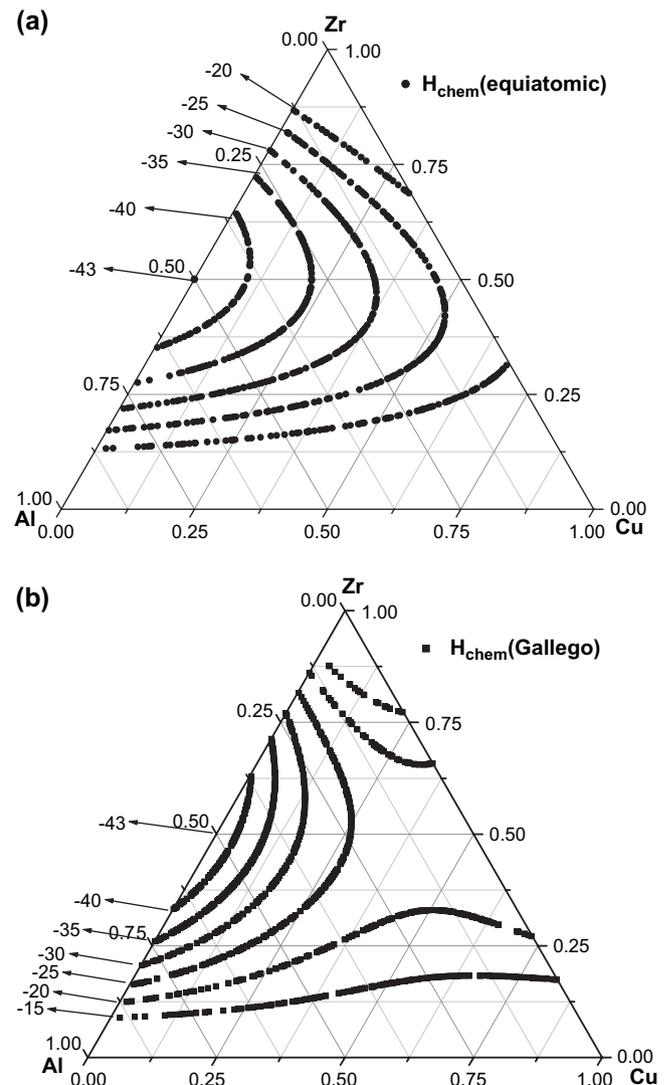


Fig. 3. Isometric contours of enthalpy due to chemical mixing, ΔH^{chem} in kJ/mol by (a) equiatomic model and (b) Gallego's model.

Isometric contours for mismatch entropy normalized by Boltzmann’s constant (S_{σ}/k_B) were drawn for Zr–Cu–Al system as shown in Fig. 2. It is seen from the figure that the contour originates from Zr–Cu rich end, with maximum value of 0.27 for the binary Zr–Cu composition. This is because S_{σ}/k_B depends on atomic diameter and composition, as is evident from Eq. (6). Atomic diameters of Zr, Cu and Al are 0.320, 0.256 and 0.286 nm, respectively. The large difference between atomic diameters between Zr–Cu leads to more randomness due to atomic misfit in comparison to Zr–Al or Al–Cu due to which S_{σ}/k_B contour is found to have high value for Zr–Cu.

Similarly isometric contours of enthalpy of chemical mixing (ΔH^{chem}) using equiatomic and Gallego’s approaches are plotted in Fig. 3(a) and (b), respectively. Eqs. (1) and (2) show the ΔH^{chem} not only depends on the composition but also on the interactions among elements, due to which different type of contours are obtained for different systems. From Fig. 3(a) the highest enthalpy of mixing is seen at $\text{Zr}_{50}\text{Al}_{50}$ (–44 kJ/mol), which continuously decreases to (–23 kJ/mol) at $\text{Zr}_{50}\text{Cu}_{50}$. This can be attributed to differences in the atomic interaction between the elements, which is evident from the enthalpy values for infinite dilution limit of $\Delta H_{\text{Zr in Al}}^{\text{inter}} = -189$ kJ/mol, $\Delta H_{\text{Al in Zr}}^{\text{inter}} = -164$ kJ/mol, $\Delta H_{\text{Zr in Cu}}^{\text{inter}} = -110$ kJ/mol,

$\Delta H_{\text{Cu in Zr}}^{\text{inter}} = -78$ kJ/mol and the nature of Eq. (1) which takes into account the equiatomic approach. Similar type of isometric contours (Fig. 3(b)) were plotted by using Gallego’s approach which takes into account the exact stoichiometric chemical enthalpy contribution of each element as seen in Eqs. (3) and (4). Here the nature and enthalpy contours are similar to that of equiatomic approach at higher enthalpy side (Zr–Al) and the pattern changes significantly at lower values (Zr–Cu side).

Superimposing of S_{config}/R , ΔH^{chem} and S_{σ}/k_B contours for Zr–Cu–Al system, as shown in Fig. 4(a) and (b), can lead to interesting information at intersections between ΔH^{chem} and S_{σ}/k_B contours in the chosen S_{config}/R regime (0.8–1.0). As mentioned before, S_{config}/R is thermodynamically a statistical parameter and depends only on the composition rather than on any intrinsic property of the elements of the alloy system. Therefore in the present study S_{config}/R is only included to locate the randomness originating by compositional configuration. On the other hand, the value of ΔH^{chem} and S_{σ}/k_B depends on intrinsic properties of elements and their interactions and the quantities of the elements present. For clarity an isometric contours of ΔH^{chem} and S_{σ}/k_B were plotted with intervals of –5 kJ/mol and 0.5, respectively. At the junctions where ΔH^{chem} and S_{σ}/k_B intersect in the chosen S_{config}/R regime,

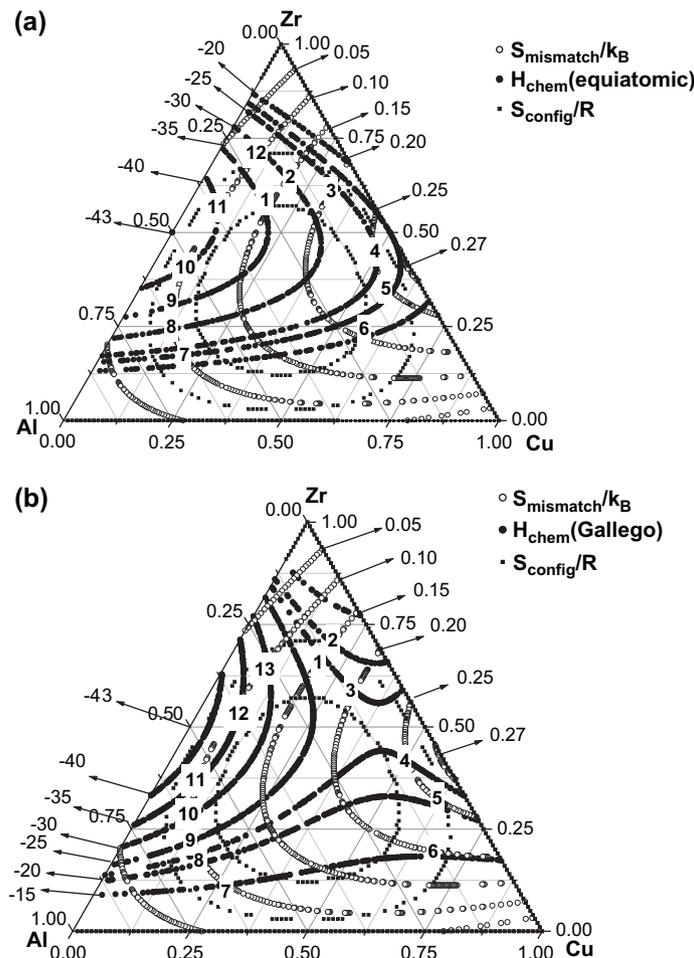


Fig. 4. Superposition of different contours showing values of $\Delta H^{\text{chem}} \times S_{\sigma}/k_B$ in kJ/mol at various junctions by (a) equiatomic model and (b) Gallego’s model.

$\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ is calculated. S_{σ}/k_{B} is a dimensionless quantity and its multiplication with ΔH^{chem} will identify compositions with high disorder combined with high negative enthalpy of chemical mixing. This confirms well with the empirical rule laid by Inoue et al. [4] for the synthesis of bulk metallic glasses from the viewpoint of enthalpy of mixing between main constituent element and disorder by multicomponent system. The product $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ for all the intersections gives interesting result, which is tabulated in Tables 3 and 4 for equiatomic and Gallego's approaches, respectively. In spite of high value of enthalpy due to chemical mixing between Zr–Al ($\Delta H_{\text{ZrAl}}^{\text{chem}}$), one can see higher values $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ product in Zr–Cu rich side in both the approaches. This is because of high values of randomness due to atomic size mismatch (S_{σ}/k_{B}) found in Zr–Cu rich compositions, which arrests the crystallization and improves the glass forming ability. Values for highest $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ differ slightly by both the approaches advocating that either of the approaches can be chosen to find out the compositions. It is obvious from the phase diagram of Zr–Cu system where metastable deep eutectic forms at equiatomic composition, which suggests that one can use equiatomic enthalpy model and the fact that Gallego's model also gives similar values of the product and similar BMG compositions strengthens this argument.

The results of the experiments conducted in the present study on all the compositions are shown in Table 1. These results have confirmed the formation of BMGs of upto 3 mm diameter in Zr–Cu rich ternary compositions, which are near to binary equiatomic Zr–Cu. Fig. 5 shows the XRD pattern of all the composition, which shows the characteristic diffraction pattern of amorphous phase. One crystalline XRD pattern is also shown for the composition of $\text{Zr}_{65}\text{Cu}_{19}\text{Al}_{16}$ as a representative of all the compositions, which could not form glassy phase in 3 mm castings. It is important to note that all the BMG compositions have a $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ value ≥ 5.0 kJ/mol, while the crystalline compositions have a lower value of the product. Interestingly, if one observes all the values of $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ on both sides of ternary diagram (Fig. 4(a) and (b)), one can find that the product values on Zr–Al side are lower in comparison to Zr–Cu side. This can explain the higher glass forming ability on Zr–Cu side in comparison to

Table 4
 $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ values by Gallego's model [16]

S. no.	Ternary map points	Compositions	ΔH^{chem} (kJ/mol)	S_{σ}/k_{B}	$\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$	Comments
1	4	$\text{Zr}_{42}\text{Cu}_{50.5}\text{Al}_{7.5}$	–23	0.25	–5.7	BMG
2	5	$\text{Zr}_{30.5}\text{Cu}_{64}\text{Al}_{5.5}$	–20	0.25	–5.0	BMG
3	3	$\text{Zr}_{58.5}\text{Cu}_{31}\text{Al}_{10.5}$	–23	0.20	–4.6	BMG
4	11	$\text{Zr}_{37}\text{Cu}_8\text{Al}_{55}$	–35	0.10	–3.5	Non-BMG
5	12	$\text{Zr}_{56.5}\text{Cu}_{7.5}\text{Al}_{36}$	–35	0.10	–3.5	Non-BMG
6	1	$\text{Zr}_{65}\text{Cu}_{19}\text{Al}_{16}$	–23	0.15	–3.5	Non-BMG
7	6	$\text{Zr}_{19}\text{Cu}_{68.5}\text{Al}_{12.5}$	–15	0.20	–3.0	Non-BMG
8	2	$\text{Zr}_{70}\text{Cu}_{20}\text{Al}_{10}$	–20	0.15	–3.0	Non-BMG
9	13	$\text{Zr}_{66}\text{Cu}_{8.5}\text{Al}_{25.5}$	–30	0.10	–3.0	Non-BMG
10	10	$\text{Zr}_{27}\text{Cu}_{10}\text{Al}_{63}$	–30	0.10	–3	Non-BMG
11	9	$\text{Zr}_{22}\text{Cu}_{14}\text{Al}_{64}$	–25	0.10	–2.5	Non-BMG
12	8	$\text{Zr}_{17}\text{Cu}_{20}\text{Al}_{64}$	–20	0.10	–2	Non-BMG
13	7	$\text{Zr}_{11.5}\text{Cu}_{27}\text{Al}_{61.5}$	–15	0.10	–1.5	Non-BMG

Zr–Al side. Very importantly there appears to be only a little difference between $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ values between crystalline (intermetallic) and amorphous compositions as shown in Tables 1, 3 and 4. Minor fluctuations in chemical composition can lead to amorphous \leftrightarrow intermetallic transitions as enthalpy difference between them is expected to be small. This is in agreement with Wang et al. [14] who also observed that small fluctuations in chemical composition can change the kinetics of amorphous phase formation.

5. Conclusions

Best glass forming composition have been successfully identified for Zr–Cu–Al system with help of contours of different thermodynamic parameters, namely, enthalpy of chemical mixing (ΔH^{chem}) by equiatomic and Gallego's models, the mismatch entropy normalized by Boltzmann's constant (S_{σ}/k_{B}) and configuration entropy S_{config}/R . The product $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ has good relation with bulk glass formation in the S_{config}/R range of 0.8–1.0. Values of the products obtained by both the models were found to be similar,

Table 3
 $\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$ values by equiatomic model [10]

S. no.	Ternary map Points	Compositions	ΔH^{chem} (kJ/mol)	S_{σ}/k_{B}	$\Delta H^{\text{chem}} \times S_{\sigma}/k_{\text{B}}$	Comments
1	4	$\text{Zr}_{45}\text{Cu}_{49}\text{Al}_6$	–25	0.25	–6.2	BMG
2	5	$\text{Zr}_{36}\text{Cu}_{58}\text{Al}_6$	–23	0.25	–5.7	BMG
3	1	$\text{Zr}_{57}\text{Cu}_{18}\text{Al}_{25}$	–35	0.15	–5.2	Non-BMG
4	3	$\text{Zr}_{61}\text{Cu}_{32}\text{Al}_7$	–25	0.20	–5.0	BMG
5	2	$\text{Zr}_{64}\text{Cu}_{20}\text{Al}_{16}$	–30	0.15	–4.5	Non-BMG
6	6	$\text{Zr}_{22}\text{Cu}_{58}\text{Al}_{20}$	–20	0.20	–4.0	Non-BMG
7	11	$\text{Zr}_{57}\text{Cu}_7\text{Al}_{36}$	–40	0.10	–4.0	Non-BMG
8	10	$\text{Zr}_{41}\text{Cu}_8\text{Al}_{51}$	–40	0.10	–4.0	Non-BMG
9	9	$\text{Zr}_{31}\text{Cu}_{10}\text{Al}_{59}$	–35	0.10	–3.5	Non-BMG
10	12	$\text{Zr}_{66}\text{Cu}_{8.5}\text{Al}_{25.5}$	–35	0.10	–3.5	Non-BMG
11	8	$\text{Zr}_{24.5}\text{Cu}_{13}\text{Al}_{62.5}$	–30	0.10	–3.0	Non-BMG
12	7	$\text{Zr}_{15}\text{Cu}_{21.5}\text{Al}_{63.5}$	–20	0.10	–2.0	Non-BMG

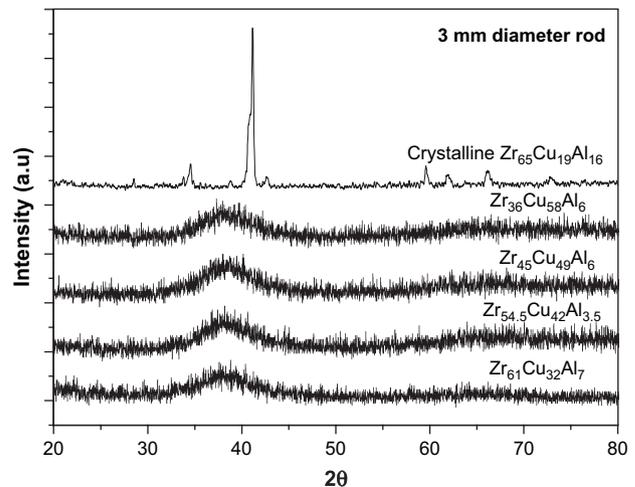


Fig. 5. XRD patterns of 3 mm diameter rods of different Zr–Cu–Al compositions.

suggesting that either of them can be used to find out the BMG compositions.

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References

- [1] Klement W, Willens RH, Duwez P. *Nature* 1960;187:869.
- [2] Chen HS. *Acta Metall* 1974;22:1505.
- [3] Johnson WL. *Prog Mater Sci* 1986;30:81.
- [4] Inoue A, Zhang T, Masumoto T. *Mater Trans JIM* 1989;30:965.
- [5] Turnbull D. *Contemp Phys* 1969;10:473.
- [6] Lu ZP, Liu CT. *Acta Mater* 2002;50:3501.
- [7] Mondal K, Murty BS. *J Non-Cryst Solids* 2005;351:1366.
- [8] Mondal K, Chatterjee UK, Murty BS. *Appl Phys Lett* 2003;83:671.
- [9] Fecht HJ, Johnson WL. *Mater Sci Eng A* 2004;375–377:2.
- [10] Takeuchi A, Inoue A. *Mater Trans JIM* 2000;41:1372.
- [11] Busch R, Johnson WL. *Appl Phys Lett* 1998;72:2965.
- [12] Murty BS, Ranganathan S, Mohan Rao M. *Mater Sci Eng A* 1992; 149:231.
- [13] Liu CT, Lu ZP. *Intermetallics* 2005;13:415.
- [14] Wang D, Li Y, Sun BB, Sui ML, Lu K, Ma E. *Appl Phys Lett* 2004; 84:4029.
- [15] Wang Q, Yang YM, Qiang JB, Zhang XF, Shek CH, Dong C. *Intermetallics* 2004;12:1229.
- [16] Gallego LJ, Somoga JA, Alonso JA. *J Phys Condens Matter* 1990;2:6245.
- [17] Niessen AK, deBoer FR, Boom R, deChatel PF, Mattens WCM, Miedema AR. *Calphad* 1983;7:51.
- [18] Mansoori GA, Carnahan NF, Startling KE, Leland Jr TW. *J Chem Phys* 1971;54:1523.
- [19] Smithells CJ, Brandes EA. *Metals reference book*. 5th ed. London: Butterworths; 1976. pp. 100–102.
- [20] Li Y, Liu HY, Jones H. *J Mater Sci* 1996;31:1957.
- [21] Inoue A, Nakamura T, Nishiyama N, Masumoto T. *Mater Trans JIM* 1992;33:937.
- [22] Inoue A, Nakamura T, Sugita T, Zhang T, Masumoto T. *Mater Trans JIM* 1993;34:351.
- [23] Tan H, Lu ZP, Yao HB, Yao B, Feng YP, Li Y. *Mater Trans JIM* 2001;42:551.
- [24] Steinberg J, Lord AE, Lacy LL, Johnson WL. *Appl Phys Lett* 1981; 38:135.
- [25] Matey JR, Anderson AC. *J Non-Cryst Solids* 1977;23:129.
- [26] Inoue A, Zhang W, Zhang T, Kurosaka K. *Acta Mater* 2001;29:2645.
- [27] Concustell A, Alcalá G, Mato S, Woodcock TG, Gebert A, Eckert J, et al. *Intermetallics* 2005;13:1214.
- [28] Yu P, Bai HY, Tang MB, Wang WL. *J Non-Cryst Solids* 2005;351:1328.
- [29] Wang YM, Shek CH, Qiang JB, Wong CH, Chen WR, Dong C. *Scripta Mater* 2003;48:1525.
- [30] Yingmin Wang, Xinfang Zhang, Jianbing Qiang, Qing Wang, Dehe Wang, Dejun Li, et al. *Scripta Mater* 2004;50:829.
- [31] Wei YX, Xi XK, Zhao DQ, Pan MX, Wang WH. *Mater Lett* 2005; 59:945.