## **Evaluation on the reliability of criterions for glass-forming ability of bulk metallic glasses**

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In recent years, there have been increasing interests in developing bulk metallic glasses (BMG) with great glass-forming ability (GFA) owing to the unique properties different from that of conventional materials and the potential engineering applications  $[1-3]$  $[1-3]$ . GFA, an important factor related to the ease of transition and a foundation for designing new BMGs, can be evaluated by means of the critical cooling rate  $(R_c)$ , which is the minimum cooling rate to maintain the melt amorphous without precipitation of crystals during solidification [\[4,](#page-4-2) [5\]](#page-4-3). In other words,  $R_c$  is the cooling rate bypassing the nose of the continuous-cooling-transition (CCT) or time-temperature-transition (TTT) curve. Lower  $R_c$ always corresponds to higher GFA [\[6\]](#page-4-4).

However, it is very difficult to measure the actual cooling rate precisely due to some uncertain factors, such as temperature variations, heterogeneous nucleation, etc. [\[7,](#page-4-5) [8\]](#page-4-6). Therefore, a great deal of effort has been focused on the researching for a reliable indicator. As a result, some parameters have been suggested to reflect the GFA of bulk amorphous alloys [\[9–](#page-4-7)[12\]](#page-4-8). For example, the temperature interval of the supercooled liquid region  $\Delta T_{\text{xg}}$ , the reduced glass transition temperature  $T_{\text{rg}}$ , the stability parameter *S* and parameters  $K_{gl}$  and  $\gamma$  have been suggested to evaluate the glassforming ability of bulk amorphous alloys.  $\Delta T_{\text{xg}}( = T_{\text{x}})$  $-T<sub>g</sub>$ ) is the temperature difference between the onset crystallization temperature  $T_x$  and the glass transition temperature  $T_g$  [\[9\]](#page-4-7).  $T_{rg} = T_g/T_l$  is the ratio of  $T_g$  to the liquidus temperature  $\overline{T}_1$  [\[10\]](#page-4-9). The stability parameter *S*(  $=(T_p - T_x)(T_x - T_g)/T_g)$  reported by Saad and Poulain [\[11\]](#page-4-10) is the ratio of  $(T_p - T_x)(T_x - T_g)$  to  $T_g$ , where  $T_p$ is the crystallization peak temperature. This parameter reflects the combined effect of the difference between  $T_p$  and  $T_x$ , as well as the position of glass transition and crystallization exotherm.  $K_{gl} = (T_x - T_g)/(T_m -$ *T*x)), proposed by Hruby [\[12\]](#page-4-8) based on the concept that the thermal stability of a glass on subsequent reheating is directly proportional to the ease of its formation, is the ratio of  $T_x - T_g$  to  $T_m - T_x$ , where  $T_m$  is the melting point. Lu *et al.* [\[13\]](#page-4-11) suggested the parameter  $\gamma$  ( =  $T_x/(T_g+T_1)$ , the ratio of  $T_x$  to  $T_g+T_1$ ), as a criterion for BMGs. This indicator was proposed based on the consideration of crystallization process in course of cooling and heating of the supercooled liquid.

It has been confirmed that several criterions show unobvious correlations with the GFA for BMGs or strong dependence only appears in certain BMG system. For example, the unclear relationship of  $\Delta T_{\text{xg}}$  with GFA has

been confirmed in Zr-Ti-Cu-Ni-Be alloys by Waniuk *et al.* [\[14\]](#page-4-12). This research evaluates the reliability of three extensively employed indicators, the temperature interval of the supercooled liquid region  $\Delta T_{\text{xg}}$ , the reduced glass transition temperature  $T_{rg}$  and the parameter  $\gamma$ , by comparing the regressive results corresponding to the critical cooling rate  $R_c$  or the critical section thickness *Z*<sup>c</sup> for several BMG systems including Zr-, La-, Mg-, Pd- and Ti-based alloys.

Table [I](#page-1-0) lists  $T_g$ ,  $T_x$ , and  $T_1$  for Zr-[2,3,15], La-[2,3,16], Mg-[2,3], Pd-[2,3,17] and Ti-based [\[18,](#page-4-1) [19\]](#page-4-13) bulk amorphous alloys. All of the data for Zr-, La-, Mg-, and Pd-based were obtained by differential scanning calorimetry (DSC) and differential thermal analysis (DTA) at a heating rate of 0.333 K/s and that for Ti-based alloys were measured by DSC at a heating rate of 0.667 K/s and DTA at a heating rate of 0.333 K/s  $[18, 18]$  $[18, 18]$ [19\]](#page-4-13). Same heating rate for measurements is emphasized due to the strong dependence of these characteristic parameters on it.

GFA can be evaluated by means of the critical cooling rate  $R_c$ , but  $R_c$  for a melt is very difficult to measure. The critical cooling rates for recently developed Zr-, La-, Mg- and Pd-based BMGs are utilized as a reflection of GFA based on corresponding references. Sometimes, the critical section thickness  $Z_c$  is also regarded as the embodiment of GFA, but this parameter usually shows less reliable than the critical cooling rate because it is very sensitive to different manufacturing techniques, such as water quenching, suction casting, high-pressure die casting, etc. Here,  $Z_c$  working as a reflection of GFA for Ti-based BMGs contributes to three aspects: (1) All of the Ti-based BMGs are prepared by arc melting high purity pre-mixed alloys, followed by ejecting into a Cu metallic mold. (2) The parameters associated with glass transition and crystallization for these injection-cast alloys are measured by DSC at a heating rate of 0.667 K/s and DTA at a heating rate of 0.333 K/s. (3) The critical cooling rate for these alloys cannot be concluded based on present researches.

In addition, the values of three criterions,  $\Delta T_{\text{xg}}$ ,  $T_{\text{rg}}$ and  $\gamma$ , have been calculated referring to above data and are also summarized in Table [I.](#page-1-0)

To reveal the relationship between indicators and the critical cooling rate  $R_c$  or the critical section thickness  $Z_c$ , the regression line for each alloy system is established respectively according to the data listed in Table [I.](#page-1-0) The regression line is expressed as a function of  $log_{10}Z_c$  or  $log_{10}R_c$  with the criterion and the

<span id="page-1-0"></span>TABLE I The glass transition temperature  $T_g$ , crystallization temperature  $T_x$ , liquidus temperature  $T_1$ , as well as the GFA criterions  $\Delta T_{xg}$ ,  $T_{rg}$ and  $\gamma$ .

	$T_{\rm g}$ (K)	$T_{\rm x}$ (K)	$T_1$ (K)	$\Delta T_{\text{Xg}}$ (K)	$T_{\rm rg}$	$\gamma$	$R_c(K/s)$
$Zr_{66}Al_8Ni_{26}$	672	707.6	1251	35.6	0.5372	0.3680	66.6 [20]
$Zr_{66}Al_8Cu_7Ni_{19}$	662.3	720.7	1200.8	58.4	0.5515	0.3868	22.7 [20]
$Zr_{66}Al_8Cu_{12}Ni_{14}$	655.1	732.5	1172.1	77.4	0.5589	0.4009	9.8 [20]
Zr <sub>66</sub> Al <sub>9</sub> Cu <sub>16</sub> Ni <sub>9</sub>	657.2	736.7	1170.6	79.5	0.5614	0.4031	$4.1$ [20]
$Zr_{57}Ti_{5}Al_{10}Cu_{20}Ni_{8}$	676.7	720	1145.2	43.3	0.5909	0.3952	10[21]
Zr <sub>38.5</sub> Ti <sub>16.5</sub> Ni <sub>9.75</sub> Cu <sub>15.25</sub> Be <sub>20</sub>	630	678	1003	48	0.6281	0.4152	$1.4$ [15]
Zr <sub>39.88</sub> Ti <sub>15.12</sub> Ni <sub>9.98</sub> Cu <sub>13.77</sub> Be <sub>21.25</sub>	629	686	1006	57	0.6252	0.4196	$1.4$ [15]
$Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$	625	739	1206	114	0.5182	0.4036	12.5 [15]
Zr <sub>45.38</sub> Ti <sub>9.62</sub> Cu <sub>8.75</sub> Ni <sub>10</sub> Be <sub>26.25</sub>	623	740	1239	117	0.5028	0.3974	17.5 [15]
$La5Al25Ni20$	490.8	555.1	941.3	64.3	0.5214	0.3876	67.5 [22]
$La55Al25Ni15Cu5$	473.6	541.2	899.6	67.6	0.5265	0.3941	34.5 [22]
$La55Al25Ni10Cu10$	467.4	547.2	835	79.8	0.5598	0.4201	22.5 [22]
$La55Al25Ni5Cu15$	459.1	520	878.1	60.9	0.5228	0.3889	35.9 [22]
$La55Al25Cu20$	455.9	494.8	896.1	38.9	0.5088	0.3660	72.3 [22]
$La55Al25Cu10Ni5Co5$	465.2	541.8	822.5	76.6	0.5656	0.4208	18.8 [22]
$La66Al14Cu20$	395	449	731	54	0.5404	0.3988	37.5 [16]
$Mg_{80}Ni_{10}Nd_{10}$	454.2	470.5	878	16.3	0.5173	0.3532	1251.4 [23]
$Mg_{75}Ni_{15}Nd_{10}$	450	470.4	789.8	20.4	0.5698	0.3794	46.1 [23]
$Mg_{70}Ni_{15}Nd_{15}$	467.1	489.4	844.3	22.3	0.5532	0.3732	178.2 [23]
$Mg_{65}Ni_{20}Nd_{15}$	459.3	501.4	804.9	42.1	0.5706	0.3966	30 [24]
$Mg_{65}Cu_{25}Y_{10}$	424.5	479.4	770.9	54.9	0.5507	0.4010	50 [25]
$Pd_{40}Cu_{30}Ni_{10}P_{20}$	576.9	655.8	836	78.9	0.6901	0.4642	$0.1$ [26]
Pd <sub>79.5</sub> Cu <sub>4</sub> Si <sub>16.5</sub>	635	675	1086	40	0.5847	0.3922	500 [27]
$Pd_{77}$ $\zeta Cu_6Si_{16}$ $\zeta$	637	678	1058.1	41	0.6020	0.4000	100 [28]
$Pd_{77}Cu_{6}Si_{17}$	642.4	686.4	1128.4	44	0.5693	0.3876	125 [29]
$\mathrm{Pd}_{40}\mathrm{Ni}_{40}\mathrm{P}_{20}$	590	671	991	81	0.5954	0.4244	$0.167$ [30]
$Ti_{50}Ni_{15}Cu_{32}Sn_{3}$	686	759	1283	73	0.57	0.4023	$1 [19]$ <sup>a</sup>
Ti <sub>50</sub> Ni <sub>15</sub> Cu <sub>25</sub> Sn <sub>3</sub> Be <sub>7</sub>	688	733	1207	45	0.61	0.4021	$2 [18]^{a}$
$Ti_{40}Zr_{25}Ni_8cu_9Be_{18}$	621	668	1009	47	0.66	0.4313	8 [19] <sup>a</sup>
$Ti_{49}Ni_{15}Cu_{25}Sn_3Be_7Zr_1$	685	733	1207	48	0.61	0.4015	$2 [18]^{a}$
$Ti_{47}Ni_{15}Cu_{25}Sn_3Be_7Zr_3$	687	741	1160	54	0.64	0.4213	$3 [18]^{a}$
$Ti45Ni15Cu25Sn3Be7Zr5$	685	741	1142	56	0.65	0.4219	$5 [18]^{a}$
$Ti_{43}Ni_{15}Cu_{25}Sn_3Be_7Zr_7$	689	743	1142	54	0.65	0.4227	$4 [18]^{a}$

The parameters for Zr-, La-, Mg- and Pd-based alloys were obtained by DSC and DTA at a heating rate of 0.333 K/s and those for Ti-based alloys were measured by DSC at a heating rate of 0.667 K/s and DTA at a heating rate of 0.333 K/s.

<span id="page-1-1"></span><sup>a</sup>Critical section thickness  $Z_c$  (mm) from the Ti-based BMGs fabricated by arc melting/copper mold injection casting



*Figure 1* The correlations between the criterions *T*xg, *T*rg and γ versus the critical cooling rate *R*<sup>c</sup> for Zr-based  $\overline{BMGs}$ : 1-Z $r_{66}A\overline{l}_8Ni_{26}$ ; 2-Z $r_{66}A\overline{l}_8Cu_7Ni_{19}$ ; 3-Z $r_{66}A\overline{l}_8Cu_{12}Ni_{14}$ ; 4-Z $r_{66}\overline{A}\overline{l}_9Cu_{16}Ni_{9}$ ; 5-Z $r_{57}Ti_{5}Al_{10}Cu_{20}Ni_{8}$ ; 6-Z $r_{38.5}Ti_{16.5}Ni_{9.75}Cu_{15.25}Be_{20}$ ; 7-Zr39.88Ti15.12Ni9.98Cu13.77Be21.25; 8-Zr44Ti11Cu10Ni10Be25; 9-Zr45.38Ti9.62Cu8.75Ni10Be26.25.

reliability of the fit is evaluated by the statistical correlation parameter  $R^2$ .  $R^2$  is a parameter in the range of 0–1 and can be obtained by using a regression program. Higher  $R^2$  indicates higher reliability of the fit. In addition, the predicted error band, a narrower interval implying lower scatter of the data and a more reliable correlation between the parameters, is shown as two dashed lines in each figure at a fixed confidence level of 95%.

Fig. [1](#page-1-1) shows the correlations between three criterions and the critical cooling rate  $R_c$  for Zr-based BMGs. Among them, the parameter  $\gamma$  shows a high  $R^2$  of 0.87 and a narrow predicted error band, which indicates that there is a strong correlation between  $R_c$  and the

<span id="page-2-0"></span>

<span id="page-2-1"></span>*Figure 2* Regressive results based on the relationships between the criterions  $\Delta T_{\text{xp}}$ ,  $T_{\text{rp}}$  and  $\gamma$  and the critical cooling rate  $R_c$  for La-based BMGs; 1-La<sub>55</sub>Al<sub>25</sub>Ni<sub>20</sub>; 2-La<sub>55</sub>Al<sub>25</sub>Ni<sub>15</sub>Cu<sub>5</sub>; 3-La<sub>55</sub>Al<sub>25</sub>Ni<sub>10</sub>Cu<sub>10</sub>; 4-La<sub>55</sub>Al<sub>25</sub>Ni<sub>5</sub>Cu<sub>15</sub>; 5-La<sub>55</sub>Al<sub>25</sub>Cu<sub>20</sub>; 6-La<sub>55</sub>Al<sub>25</sub>Cu<sub>10</sub>Ni<sub>5</sub>Co<sub>5</sub>; 7-La<sub>66</sub>Al<sub>14</sub>Cu<sub>20.</sub>



<span id="page-2-2"></span>*Figure 3* The correlations between the criterions  $\Delta T_{\text{xg}}$ ,  $T_{\text{rg}}$  and  $\gamma$  versus the critical cooling rate  $R_c$  for Mg-based BMGs; 1-Mg<sub>80</sub>Ni<sub>10</sub>Nd<sub>10</sub>; 2-Mg<sub>75</sub>Ni<sub>15</sub>Nd<sub>10</sub>; 3-Mg<sub>70</sub>Ni<sub>15</sub>Nd<sub>15</sub>; 4-Mg<sub>65</sub>Ni<sub>20</sub>Nd<sub>15</sub>; 5-Mg<sub>65</sub>Cu<sub>25</sub>Y<sub>10.</sub>



*Figure 4* Regressive results based on the relationships between the criterions  $\Delta T_{\text{xg}}$ ,  $T_{\text{rg}}$  and  $\gamma$  and the critical cooling rate  $R_c$  for Pd-based BMGs; 1-Pd40Cu30Ni10P20; 2-Pd79.5Cu4Si16.5; 3-Pd77.5Cu6Si16.5; 4-Pd77Cu6Si17; 5-Pd40Ni40P20.

parameter  $\gamma$  for Zr-based BMGs. Therefore,  $\gamma$  shows the highest reliability for selected Zr-based alloys. Furthermore, the indicator  $\Delta T_{\text{xg}}$  exhibits a very low  $R^2$  of 0.05 and a wide predicted error band, which is characterized in the scattered date. It indicates that no obvious correlation between  $\Delta T_{\text{xg}}$  and  $R_c$ , hence the lowest reliability of  $\Delta T_{\text{xg}}$  for chosen Zr-based alloys.  $T_{\text{rg}}$  shows a moderate  $R^2$  of 0.62. So, the reliability of these indicators for Zr-based BMGs listed in Table [I](#page-1-0) can be summarized as:  $\Delta T_{\text{xg}} \ll T_{\text{rg}} \ll \gamma$ .

For La-based BMGs, as shown in Fig. [2,](#page-2-0)  $\Delta T_{\text{xg}}$  shows the lowest  $R^2$  of 0.63, while  $R^2$  values for  $T_{\text{rg}}$  and  $\gamma$  are 0.82 and 0.85, respectively, which implies that both  $T_{\text{rg}}$  and  $\gamma$  have nearly equal reliabilities for La-based system. Accordingly, following relationship can be satisfied:  $\Delta T_{\text{xg}} < T_{\text{rg}} \gamma$ . Also, similar rule can be concluded

<span id="page-3-0"></span>

*Figure 5* Regressive results based on the relationships between the criterions *T*xg, *T*rg and γ and the critical section thickness *Z*<sup>c</sup> for Td-based BMGs; 1-Ti50Ni15Cu32Sn3; 2-Ti50Ni15Cu25Sn3Be7; 3-Ti40Zr25Ni8Cu9Be18; 4-Ti49Ni15Cu25Sn3Be7Zr1; 5-Ti47Ni15Cu25Sn3Be7Zr3; 6- Ti45Ni15Cu25Sn3Be7Zr5; 7-Ti43Ni15Cu25Sn3Be7Zr7.

for Mg-based alloys, as shown in Fig. [3,](#page-2-1) in which the values of  $R^2$  are 0.40, 0.87 and 0.83 for  $\Delta T_{\text{xg}}$ ,  $T_{\text{rg}}$  and  $\gamma$ , respectively.

The regression for Pd-based BMGs demonstrates a high  $R^2$  value of 0.98 for  $\Delta T_{\text{xg}}$ , while a considerably low one of 0.48 for  $T_{rg}$ , which is entirely different from the trends for Zr-, La- and Mg-based systems. In addition, a moderate  $R^2$  value of 0.81 for the parameter  $\gamma$ , as shown in Fig. [4.](#page-2-2) The reliability of these criterions for Pd-based BMGs can be expressed as:  $T_{rg} < \gamma < \Delta T_{xg}$ .

Fig. [5](#page-3-0) shows the correlations between selected three criterions and the critical section thickness  $Z_c$  for Tibased BMGs. Here,  $Z_c$  instead of the critical cooling rate  $R_c$  is considered as the reflection of GFA due to the lack of data related to  $R_c$ . Among them, the parameter  $T_{\text{rg}}$  shows the highest  $R^2$  of 0.93, hence a strong correlation between  $Z_c$  and the criterion for Ti-based BMGs. The indicator  $\Delta T_{\text{xg}}$  exhibits a very low  $R^2$  of 0.20 and a wide predicted error band, which indicates that no obvious correlation between  $\Delta T_{\text{xg}}$  and  $Z_c$  for chosen Ti-based alloys. Indicator  $\gamma$  shows a moderate  $R^2$  of 0.86 that is a little lower than that of  $T_{\text{re}}$ .

<span id="page-3-1"></span>

*Figure 6* The statistical correlation parameter  $R^2$  showing as a function of bulk amorphous alloys including Zr-, La-, Mg-, Pd- and Ti-based systems.

Therefore, following expression can be concluded for Ti-based BMGs:  $\Delta T_{\text{xg}} \ll \gamma \ll T_{\text{rg}}$ . In addition, it can be seen that the correlation of the indicators with  $Z_c$  for Tibased BMGs is quite similar to that with  $R_c$  for above systems except for Pd-based alloys.

According to the results calculated above, the statistical correlation parameter  $R^2$  is shown as a function of BMGs including Zr-, La-, Mg-, Pd- and Ti-based systems, referring to Fig. [6.](#page-3-1) The temperature interval  $\Delta T_{\text{xe}}$  shows weak correlations with all alloy systems except Pd-based BMGs. It is known that GFA represents the ease that melts are cooled to form amorphous alloys without any crystallization [\[6\]](#page-4-4). Generally, a large  $\Delta T_{\text{xg}}$  value implies that the supercooled liquid can exist in a wide temperature range without crystallization and has a high resistance to the nucleation and growth of crystalline phases, leading to good GFA. However,  $\Delta T_{\text{xg}}$  is a parameter for the estimation of glass stability which is defined as the resistance of glasses towards devitrification upon reheating above the glass transition temperature  $T_g$ . In fact, an increasing GFA does not always lead to an enhanced stability. Hence it is inappropriate to utilize  $\Delta T_{\text{xg}}$  as a criterion of GFA. For chosen Ti-based BMGs, the relationship between  $\Delta T_{\text{xe}}$ and  $Z_c$  actually shows a contrary tendency compared with the theoretical analysis.

The criterion  $T_{rg}$  is proposed on the basis of kinetic reasons related to the cooling process without any crystallization, which is on the assumption that the liquidus temperature  $T_1$  decreases obviously with the alloy composition, while the glass transition temperature  $T_g$  is less dependent. As a result,  $T_{rg}$  increases with the increase in alloying concentration, thus an increasing GFA. This is more suitable for binary systems instead of multicomponent systems owing to the great variations in  $T_g$  and  $T_1$ . In addition, high viscosity of glasses in the range of temperatures  $T_g$  and  $T_l$  is essential condition for an increasing GFA, but the constant viscosity of  $10^{12}$  Pa s at  $T_g$  combined with the different variation of temperature with viscosity for different systems makes  $T_1$  reflect GFA alone, thus leads to large error for some multicomponent systems. The analysis above interprets that  $T_{\text{rg}}$  shows low  $R^2$  of 0.62 and 0.48

for Zr- and Pd-based systems, respectively, while a high value of 0.93 for Ti-based BMGs.

The parameter  $\gamma$  has been proposed as a criterion to reflect GFA based on the consideration of crystallization processes in course of cooling and reheating of the supercooled liquid. From this point of view, two factors, the stability of liquid phase and the resistance to crystallization, should be considered for GFA. GFA is proportional to the factor  $T_x/T_g$  on the basis of the glass stability upon the reheating process of a glass. On the other hand, the critical cooling rate  $R_c$  becomes lower with an larger  $T_x/T_1$ , which increases with increasing viscosity of the supercooled liquid, fusion entropy, activation energy of viscous flow and with decreasing liquid temperature  $T_1$  [\[13\]](#page-4-11). A series of strong correlations of this parameter with the critical cooling rate  $R_c$  have been confirmed for typical metallic glasses (bulk metallic glasses and conventional amorphous alloys)  $[13]$ , some glassy oxides  $[31]$ , as well as some cryo-protective aqueous solutions [\[31\]](#page-5-5). For these regressions, the values of the statistical correlation parameter  $R^2$  are 0.9, 0.8 and 0.9, respectively. In addition, a linear relationship has been observed between  $\gamma$  and *R*<sup>c</sup> for conventional amorphous alloys and BMGs and is expressed in an approximation formula [\[13\]](#page-4-11):

$$
R_{\rm c} = 5.1 \times 10^{21} \exp(-117.19\gamma) \tag{1}
$$

The equation can be utilized to estimate  $R_c$  when  $\gamma$  has been obtained from DSC/DTA measurements, which is significant for the evaluation of GFA for new developed BMGs. Also, several equations that indicate the relationship between  $R_c$  and  $\gamma$  have been obtained for selected BMG systems. Although none of the *R2* values is higher than that from Ref. [\[13\]](#page-4-11), the new parameter shows the best reliability among the three indicators. High *R2* value of 0.91 obtained by Lu *et al.* is based on the consideration of metal glasses including conventional amorphous alloys and BMGs. Generally, non-BMGs show higher  $R_c$  value than BMGs and a larger difference in  $R_c$  value results in a higher  $R^2$ value, such as the maximum  $R_c$  values of  $3.0 \times 10^{10}$  K/s for pure Ni among selected conventional alloys and 1251.4 K/s for  $Mg_{80}Ni_{10}Nd_{10}$  BMG [\[13\]](#page-4-11). In this research, the maximum difference is around 1220 K/s between  $Mg_{80}Ni_{10}Nd_{10}$  and  $Mg_{65}Ni_{25}Nd_{10}$ , thus corresponds to a lower  $R^2$  value.

The three criterions were proposed based on different considerations regarding the GFA of glasses. Present researches sufficiently demonstrate that  $\Delta T_{\text{xg}}$ is a quantitative measure or a reflection of glass stability instead of a cause of GFA due to the weak dependence of  $R_c$  or  $Z_c$  on it. Regressive result of the criterion  $T_{rg}$ with  $R_c$  for chosen metallic glasses indicates that  $R_c$  is somewhat dependent on  $T_{\text{rg}}$  with a  $R^2$  value of 0.74. It is clear that  $\gamma$  has a better correlation with  $R_c$  than  $T_{\text{rg}}$ . Similar trend can also be found for  $Z_c$ , which is characterized in a  $R^2$  value of 0.57 for  $\gamma - Z_c$  and 0.32 for  $T_{\text{rg}}$ −*Z*<sub>c</sub>. Although  $\gamma$  shows the highest reliability for all metallic glasses, it doesn't mean a high one for certain system. This research has confirmed the well correlations of the new indicator with chosen systems. For all systems, the parameter  $\gamma$  correlates well with  $Z_c$  or  $R_c$  owing to an average  $R^2$  value higher than 0.8, even though none of them is higher than 0.9. Therefore, it can be concluded that the criterion  $\gamma$  is preferred to be a universal criterion of GFA for glasses.

By comparing the regressive results of three criterions,  $\Delta T_{\text{xg}}$ ,  $T_{\text{rg}}$  and  $\gamma$ , with  $R_c$  for Zr-, La-, Mg- and Pd-based BMGs and  $Z_c$  for Ti-based alloys, the parameter  $\gamma$  correlates best with all selected systems evaluated by an average value of statistical correlation parameter  $R^2$  higher than 0.8, which indicates that  $\gamma$  is the most reliable criterion for BMGs.

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## **References**

- 1. M. OUCHETTO, B. ELOUADI and <sup>S</sup> . PARKE, *Phys. Chem. Glasses.* **32** (1991) 22.
- <span id="page-4-0"></span>2. Z. P. LU and C. T. LIU, *Scripta Mater*. **42** (2000) 667.
- 3. Z. P. LU and C. T. LIU, *J. Non-Cryst. Solids* **270** (2000) 103.
- <span id="page-4-1"></span>4. D. E. POLK, A. CALKA and B. C. GIESSEN, *Acta Metall.* **26** (1978) 1097.
- <span id="page-4-2"></span>5. T. ZHANG, A. INOUE and T. MASUMOTO, *Mater. Sci. Eng.* **A181/182** (1994) 131.
- <span id="page-4-3"></span>6. A. INOUE, N. NISHIYAMA, K. AMIYAM, T. ZHANG and T. MASUMOTO, *Mater. Lett.* **19** (1994) 131.
- <span id="page-4-4"></span>7. T. ZHANG and A. INOUE, *Mater. Sci. Eng.* **A304–306** (2001) 771.
- <span id="page-4-5"></span>8. Y. C. KIM, S. YI, W. T. KIM and D. H. KIM, Mater. Sci. *Forum* **360–362** (2001) 67.
- <span id="page-4-6"></span>9. M. MARCUS and D. TURNBULL, *Mater. Sci. Eng.* **23** (1976) 211.
- <span id="page-4-7"></span>10. C. V. THOMPSON, A. L. GREER and F. SPAEPEN, *Acta Metall.* **31** (1983) 1883.
- <span id="page-4-9"></span>11. M. SAAD and M. POULAIN, *Mater. Sci. Forum* **11** (1987) 19.
- <span id="page-4-10"></span>12. A. HRUBY, *Czech. J. Phys.* **22** (1972) 1187.
- <span id="page-4-11"></span><span id="page-4-8"></span>13. Z. P. LU and C. T. LIU, *Acta Mater.* **50** (2002) 3501.
- 14. T. A. WANIUK, J. SCHROERS and W. L. JOHNSON, *Appl. Phys. Lett.* **78** (2001) 1213.
- <span id="page-4-12"></span>15. A. INOUE, T. ZHANG, K. KUROSAKA and W. ZHANG, *Mater. Trans. JIM* **42** (2001) 1800.
- <span id="page-4-17"></span>16. H. TAN, Z. P. LU, H. B. YAO, B. YAO, Y. P. FENG and Y. L I, *Mater. Trans. JIM* **42** (2001) 551.
- <span id="page-4-19"></span>17. A. INOUE, N. NISHIYAMA and H. KIMURA, *Mater. Trans. JIM* **38** (1997) 179.
- 18. Y. C. KIM, D. H. BAE, W. T. KIM and D. H. KIM, *J. Non-Crystallogr. Solids* **325** (2003) 242.
- <span id="page-4-14"></span>19. Y. C. KIM, W. T. KIM and D. H. KIM, *Mater. Sci. Eng.* **A375–377** (2004) 127.
- <span id="page-4-13"></span>20. H. H. HNG, Y. LI, S. C. NG, C. K. ONG, *J. Non-Cryst*. *Solids* **208** (1996) 124.
- <span id="page-4-15"></span>21. L. Q. XING, P. OCHIN and M. HARMELIN, *Mater. Sci. Eng.* **A220** (1996) 155.
- <span id="page-4-16"></span>22. Z. P. LU, T. T. GOH, Y. LI and S. C. NG, *Acta Mater*. 47 (1999) 2215.
- <span id="page-4-18"></span>23. Y. LI, H. Y. LIU and H. JONES, *J. Mater. Sci.* 31 (1996) 1957.
- <span id="page-4-20"></span>24. Y. LI, H. Y. LIU, H. A. DAVIES and H. JONES, Mater. *Sci. Eng.* **A179/180** (1994) 628.
- <span id="page-4-22"></span><span id="page-4-21"></span>25. R. BUSCH, W. LIU and W. L. JOHNSON, *J. Appl. Phys.* **83** (1998) 4134.
- 26. N. NISHIYAMA and A. INOUE, *Mater. Trans. JIM* **38** (1997) 464.
- <span id="page-5-0"></span>27. L. F. CHUA and H. W. KUI, *J. Appl. Phys.* **84** (1998) 5993.
- <span id="page-5-1"></span>28. J. R. MATEY and A. C. ANDERSON, *J. Non-Crystallogr. Solids* **23** (1977) 129.
- <span id="page-5-3"></span><span id="page-5-2"></span>29. H. A. DAVIES, *Phys. Chem. Glasses* 17 (1976) 159.
- 30. J. STEINBERG, A. E. LORD, L. L. LACY and J. JOHN-SON, *Appl. Phys. Lett.* **38** (1981) 135.
- <span id="page-5-5"></span><span id="page-5-4"></span>31. Z. P. LU and C. T. LIU, *Intermetallics* **12** (2004) 1035.

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