**Rare Metal Materials and Engineering**
Volume 53, Issue 1, January 2024
Available online at www.rmme.ac.cn

Science Press

**Cite this article as**: Li Xiaocheng, Kou Shengzhong, Zhao Yanchun, et al. Triangle Criterion of Glass-Forming Ability and Stability for Metallic Glasses[J]. Rare Metal Materials and Engineering, 2024, 53(01): 31-37. DOI: 10.12442/j.issn.1002-185X.E20230034.

ARTICLE

# Triangle Criterion of Glass-Forming Ability and Stability for Metallic Glasses

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**Abstract:** Based on the glass-forming ability (GFA) during cooling process and the glass stability (GS) of heating process, a triangle to evaluate GFA and GS, namely Tri-FAS, with the combination of pseudo-four characteristic parameters as vertices was established. Accordingly, a GFA&GA criterion (G-FAS) was deduced as G-FAS=$T_g/T_l+T_x/T_l+T_x/T_g$ ($T_x$ is onset crystallization temperature; $T_l$ is liquid temperature; $T_g$ is glass transition temperature). Additionally, the criterion was modified based on the competitive relationship between amorphous phase and crystal phase during cooling process and the contribution of each component to the criterion: G-FAS$_m$=$T_g/(1.5T_x)+T_x/T_l+T_x/T_g$ and G-FAS$_m$′=$T_g/T_l+T_x/T_l+(T_x/T_g)^a$ ($a\approx1.5\pm0.2$). The correlation between G-FAS and critical cooling rate $R_c$ and that between G-FAS and $T_{xg}$ ($T_{xg}$ reflects the supercooled liquid region of glass, $T_{xg}=T_x/T_g$) were discussed, which could reflect GFA and GS, respectively. Through the determination results of GFA and GS of abundant metallic glasses and other glass formers, the validity of the proposed G-FAS criterion was evaluated. Results show that with respect to both GFA and GS, the G-FAS criterion is reliable in various glass former systems, showing wide applications. The proposed Tri-FAS and G-FAS criterion can provide guidance during the fabrication and application of metallic glasses.

**Key words:** metallic glasses; glass-forming ability; thermal stability; characteristic temperature; criterion

The manufacturing process and the actual application process attract much attention in material research. For the metallic glasses, glass-forming ability (GFA) in the preparation process and glass stability (GS) in the application process are important parameters in various high-tech industries, such as the aviation, aerospace, computing, and communication[1–7].

For the glass formation during cooling process, the stronger the GFA, the greater the metal glass size even at the same cooling rate, and accordingly the smaller the critical cooling rate $R_c$ (the minimum cooling rate to obtain fully amorphous solid from melts). In this case, the amorphous state can be easily formed. Therefore, $R_c$ can directly evaluate GFA. However, $R_c$ cannot be measured accurately[8]. Consequently, some simple but practical criteria based on the characteristic temperatures ($T_g$: glass transition temperature; $T_x$: onset crystallization temperature; $T_l$: liquid temperature) which are closely related to $R_c$ are proposed to characterize GFA, such as $\Delta T_x=T_x-T_g$[9–10], $T_{rg}=T_g/T_l$[11–13], $H'=(T_x-T_g)/T_g$[14], $\gamma=T_x/(T_g+T_l)$[15], $\Delta T_l=T_l-T_x$[16], $\Delta T_{rg}=(T_x-T_g)/(T_l-T_g)$[17], $\alpha=T_x/T_l$[18–19], $\beta=T_x/T_g+T_g/T_l$[19], $\delta=T_x/(T_l-T_g)$[20], $\varphi=T_{rg}(\Delta T_x/T_g)^a$[21], $\gamma_m=(2T_x-T_g)/T_l$[22], $\beta_Y=T_xT_g/(T_l-T_x)^2$[23], $\xi=T_g/T_l+\Delta T_x/T_x$[24], $\omega=T_g/T_x-2T_g/(T_g+T_l)$[25], $\theta=(T_x+T_g)/T_l[(T_x-T_g)/T_l]^{\alpha}$[26], $\omega_2=T_g/(2T_x-T_g)-T_g/T_l$[27], $\omega_J=T_l(T_l+T_x)/[T_x(T_l-T_x)]$[28], $\gamma_c=(3T_x-2T_g)/T_l$[29], $\beta'=T_g/T_x-T_g/(T_l\eta)$[30], $\omega_B=(2T_x-T_g)/(T_l+T_x)$[31], $G_p=T_g(T_x-T_g)/(T_l-T_x)^2$[32], $\chi=(T_x-T_g)/(T_l-T_x)(T_x/T_l-T_x)^a$[33], $\gamma_n=(5T_x-3T_g)/T_l$[34], $\bar{\omega}=(T_x-T_g)T_gT_x/(T_l-T_x)^3$[35], $k=T_gT_xT_l(T_x-T_g)/(T_l-T_x)^4$[36], G-FAS1=$T_x/T_l+(T_x-T_g)/(T_l-T_g)$[37–38], and $G_{T_g}=T_g/T_l+T_x/T_g+(T_x-T_g)/(T_l-T_g)$[39]. $T_l$ reflects the stability of high temperature liquid. When $T_g$ increases, the high temperature liquid can easily pass through the narrow interval $T_l-T_g$ (namely the high temperature melt supercooled liquid region, SLR$_{melt}$) under a small $R_c$ to form amorphous state[40].

During the heating process, good GS indicates the fine performance and internal structural stability of formed glass

Received date: September 12, 2023
Foundation item: National Natural Science Foundation of China (51971103); Key Research and Development Program of Gansu Province (20YF8GA052)
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during application. The formed glass with good GS can hardly be crystallized, showing a wide supercooled liquid region of glass ($SLR_{glass}$). In other words, it has longer service life than other metallic glasses do before crystallization failure. It is well known that GS measured by the wide interval $T_x-T_g$ is likely to cause large GFA. But in some specific alloy systems, GFA and GS show inconsistent relationship. Although GFA and GS are related concepts, they are not equivalent characterizations of glasses[41]. Through the quantitative evaluations of GFA and GS, it is known that the increase in GFA is usually not accompanied by the increase in GS with the same magnitude[42]. Generally, the GFA evaluation criteria attract more attention than the GS evaluation criteria do. Moreover, the proposed GS evaluation criteria are mostly related to GFA, such as $(T_x-T_g)/(T_l-T_x)$[42], $T_x-T_g$[9–10], $(T_x-T_g)/T_l$[43], $(T_x-T_g)/T_g$[14], and $T_x/T_l$[18–19].

Therefore, in this research, the GFA and GS evaluation criteria were combined through a simple triangle during both cooling and heating processes. On this basis, a criterion for GFA&GS evaluation was proposed and further modified from two aspects: the competitive relationship between amorphization and crystallization during the cooling process, and the contribution of components to the GFA&GS criterion.

## 1 Experiment

First, 65 types of metallic glasses with characteristic temperatures and $R_c$ values ($SiO_2$ was used as the best glass forming material) were collected by five measurement methods or $R_c$ estimation[44]. Various cryoprotective solutions (31 types) and glassy oxides (23 types) in Ref.[45] were used as the reliable database. Then, the linear interrelationship between $R_c$ and $T_{xg}$, which reflect GFA and GS, respectively, and the criterion $c$ based on characteristic temperatures were analyzed: $\lg R_c=A-Bc(T_g, T_x, T_l)$ and $T_{xg}=A+Bc(T_g, T_x, T_l)$, where $A$ and $B$ are fitting parameters. The linear regression equation and the coefficient of determination $R^2$ were obtained by Origin software through linear fitting.

## 2 Analysis

### 2.1 Revisit pseudo-four characteristic parameters

The close relationship among the characteristic temperatures, GFA&GS criterion from GFA aspect during cooling process, and that from GS aspect during heating process was investigated. A simple triangle structure of characteristic temperatures is used to illustrate their relationship, as shown in Fig. 1. $T_l$ reflects the liquid stability; $T_x$ reflects the resistance to crystallization; $T_g$ reflects GFA during cooling process and GS during heating process; $T_{rg}=T_g/T_l$ indicates the GFA during cooling process (from red point $T_l$ to green point $T_g$); $T_{xg}=T_x/T_g$ indicates the GS during heating process (from green point $T_g$ to brown point $T_x$); $T_{xl}=T_x/T_l$ indicates the GFA during cooling process and the GS during heating process (blue line connecting red point $T_l$ and brown point $T_x$).

Firstly, from the perspective of amorphization during cooling process (from red point $T_l$ to green point $T_g$), the

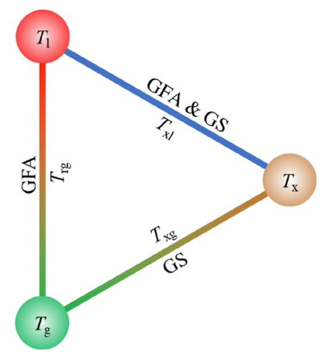


Fig.1 Triangle structure of $T_l$, $T_x$, and $T_g$ characteristic temperatures

characteristic temperature $T_l$ denotes the stability of high-temperature liquid, and the GFA or glass-forming tendency is denoted by $T_g/T_l$. The lower the $T_l$, the more stable the high-temperature liquid, i. e., equilibrium liquid can resist the solidification[46–48]. With increasing the $T_g/T_l$ ratio, the interval between $T_l$ and $T_g$ (namely $SLR_{melt}$) is decreased, so the stable high-temperature liquid can pass through the “dangerous” region without crystallization, i.e., GFA is enhanced.

Secondly, from the perspective of devitrification during heating process (from green point $T_g$ to brown point $T_x$), the characteristic temperature $T_x$ denotes the crystallization resistance, and $T_{xg}=T_x/T_g$ denotes the $SLR_{glass}$ stability. Larger $T_x$ value correlates to the higher crystallization resistance. A large $SLR_{glass}$ value suggests that the supercooled liquid of glass can exist in a wide temperature range without crystallization and presents the strong resistance against the nucleation and growth of crystalline phase, i.e., GS is enhanced[10].

Thirdly, the blue line of $T_l$ and $T_x$ ($T_{xl}=T_x/T_l$) simultaneously denotes the GFA and GS, and it is only related to $T_x$ and $T_l$ without $T_g$[19]. It is worth noting that the characteristic temperature $T_g$ is relatively special, compared with the other two characteristic temperatures $T_l$ and $T_x$. Sole $T_g$ parameter cannot provide any information about GFA&GS relationship[15]. Therefore, it can be deduced that the characteristic temperature $T_g$ reflects both GFA during cooling process and GS during heating process.

According to the abovementioned analysis, it can be seen that in the cooling process, the increase in $T_g$ can enhance GFA; whereas in the heating process, the increase in $T_x$ coupled with the decrease in $T_g$ can expand $SLR_{glass}$. Theoretically, there is a contradiction relationship between GFA&GS and $T_g$. To solve this problem, $T_g$ and $T_l$ are combined during cooling process, and $T_g$ and $T_x$ are combined during heating process, thus forming two pseudo characteristic parameters $T_g/T_l$ and $T_x/T_g$ and avoiding the sole appearance of $T_g$. Therefore, three characteristic temperatures are modified into pseudo-four characteristic parameters: $T_x$, $T_l$, $T_x/T_g$, and $T_g/T_l$. In this case, the crystallization resistance is related to $T_x$: the higher the $T_x$, the stronger the crystallization resistance of glass; the stability of high-temperature liquid is related to $T_l$: the lower the $T_l$, the more stable the liquid; the $SLR_{glass}$ stability is related to $T_x/T_g$: the larger the $T_x/T_g$, the more stable the supercooled liquid of glass; GFA/glass-forming tendency is related to $T_g/T_l$: the larger the $T_g/T_l$, the stronger the GFA of liquid.

### 2.2 Tri-FAS and G-FAS criterion

The triangle of glass-forming ability and glass stability (Tri-FAS) is established based on the pseudo-four characteristic parameters, as shown in Fig. 2. On the one hand, for GFA evaluation during cooling process and GS evaluation during heating process, three characteristic temperatures are reduced by $T_g$ to obtain $T_{rg}$ and $T_{xg}$, reduced by $T_l$ to obtain $T_{rg}$ and $T_{xl}$, and reduced by $T_x$ to obtain $T_{xg}$ and $T_{xl}$, respectively. Thus, the parameter $T_{xl}$ can reflect GFA&GS in both cooling and heating processes. On the other hand, the Tri-FAS can also be obtained by combining the pseudo-four characteristic parameters according to the principle of similar complementary in physical meaning. Among the pseudo-four characteristic parameters, the characteristic temperature $T_g$ is combined with $T_x$ and $T_l$ to obtain $T_{xg}$ and $T_{rg}$ in Tri-FAS, respectively. The remaining $T_x/T_l$ is combined with $T_x$ and $T_l$ with dual-meaning of GFA and GS. In the Tri-FAS, although $T_{xl}$ can reflect both GFA and GS[19], $T_{xl}$ cannot comprehensively characterize GFA&GS due to the lack of $T_g$. Therefore, the parts including $T_g$ should also be considered to form a unified determination criterion to evaluate GFA&GS: G-FAS=$T_g/T_l+T_x/T_l+T_x/T_g$. It should be noted that various forms can also be obtained through the combination of the pseudo-four characteristic parameters, such as $(T_g/T_l+T_x/T_g)$ $(T_x/T_l)$ (determination coefficient $R^2$=0.864), $(T_g/T_l)$ $(T_x/T_l)$ $(T_x/T_g)$ ($R^2$=0.828), and $(T_g/T_l)(T_x/T_g)+T_x/T_l$ ($R^2$=0.846). However, the proposed G-FAS criterion is the simplest and most reliable for experiment validation.

According to the classical nucleation and crystal growth theories[49–52], some expressions can be easily derived from the perspective of GFA during cooling process and GS during heating process, as follows:

$$\text{GFA}\propto T_g/T_l \tag{1}$$

$$\text{GS}\propto T_x/T_g \tag{2}$$

These two correlations form the G-FAS criterion expressed by characteristic temperature $T_g$, as follows:

$$\text{G-FAS}_{(T_g)}\propto(\text{GFA, GS}) \tag{3}$$

It is mentioned that the characteristic temperature $T_x$ in the pseudo-four characteristic parameters indicates the crystallization resistance and $T_l$ indicates the stability of high-temperature liquid. Thus, the combination of $T_x$ and $T_l$ can be used to express G-FAS criterion, as follows:

$$\text{G-FAS}_{(T_x,\,T_l)}\propto T_x/T_l \tag{4}$$

G-FAS$_{(T_g)}$ and G-FAS$_{(T_x,\,T_l)}$ can be unified, as follows:

$$\text{G-FAS}\propto(\text{G-FAS}_{(T_g)},\ \text{G-FAS}_{(T_x,\,T_l)}) \tag{5}$$

Substituting Eq.(1–4) into Eq.(5), Eq.(6) can be obtained:

$$\text{G-FAS}\propto(T_g/T_l,\ T_x/T_l,\ T_x/T_g) \tag{6}$$

Hence, the G-FAS criterion for GFA&GS evaluation is as follows:

$$\text{G-FAS}=T_g/T_l+T_x/T_l+T_x/T_g \tag{7}$$

It should be noted that the formula in Eq.(7) is not a simple superposition of three components. According to the abovementioned pseudo-four characteristic parameters, Eq.(7) should be regarded as four separate parts of $T_x$, $T_l$, $T_x/T_g$, and $T_g/T_l$. Each part has a clear physical meaning. The characteristic temperature $T_l$, which represents the stability of high-temperature liquid, is combined with $T_g$ to form the first term $T_g/T_l$, reflecting GFA/glass-forming tendency. The characteristic temperature $T_x$, which represents the crystallization resistance, is combined with $T_g$ to form the third term $T_x/T_g$, thus reflecting the SLR$_{glass}$ stability. Besides, $T_l$ and $T_x$ are combined to form the second term $T_x/T_l$, reflecting GFA and GS simultaneously.

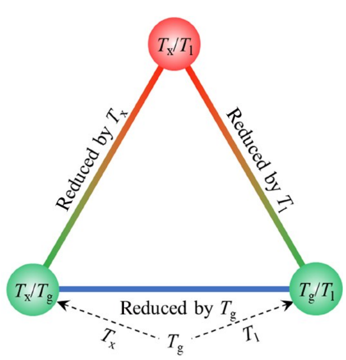


Fig.2 Schematic diagram of Tri-FAS based on pseudo-four characteristic parameters $T_x$, $T_l$, $T_{xg}$, and $T_{rg}$

### 2.3 Modification of G-FAS criterion

To further improve the reliability of the criterion for GFA&GS evaluation, the criterion is modified from the perspective of glass formation ($T_g/T_l$ term in criterion) during cooling process. In terms of glass formation, $T_g/T_l$ represents GFA: the higher the $T_g$, the easier the glass formation. On the other hand, from the perspective of metal solidification, $T_{xc}/T_l$ reflects the crystallization of high-temperature melt ($T_{xc}$ is the onset temperature of solidification): the higher the $T_{xc}$, the easier the crystal formation. Thus, from the competitive correlation between glass formation and crystallization, their ratio of $(T_g/T_l)/(T_{xc}/T_l)=T_g/T_{xc}$ jointly determines the GFA. Therefore, $T_g/T_l$ should be replaced by $T_g/T_{xc}$ in Eq. (7) to obtain the modified G-FAS criterion (G-FAS$_m$), as follows:

$$\text{G-FAS}_m=T_g/T_{xc}+T_x/T_l+T_x/T_g \tag{8}$$

According to Ref.[29], there is a linear relationship between $T_{xc}$ and $T_x$ as $T_{xc}=1.5T_x$. Considering that $T_x$ value is relatively easy to measure, Eq.(8) can also be expressed as follows:

$$\text{G-FAS}_m=T_g/(1.5T_x)+T_x/T_l+T_x/T_g \tag{9}$$

Afterwards, G-FAS criterion is further ameliorated from the perspective of the contribution of each component to the G-FAS criterion. By analyzing the $T_g/T_l$, $T_x/T_l$, and $T_x/T_g$ values of 65 metallic glasses[44], it is found that the $T_x/T_g$ values are greater than 1, whereas the $T_g/T_l$ and $T_x/T_l$ values are similar and much smaller than the $T_x/T_g$ value, such as $T_{rg}$=0.246(Ni)~(0.369~0.691)~0.726($SiO_2$), $T_{xl}$=0.246(Ni)~(0.369~0.807)~0.872($SiO_2$), and $T_{xg}$=1(Ni)~(1~1.197)~1.2($SiO_2$). The values of Ni and $SiO_2$ are lower limit and upper limit, respectively. Therefore, the $T_x/T_g$ value is exponentially revised to approach the $T_g/T_l+T_x/T_l$ value and to have the same order of magnitude.

So three terms, $T_g/T_l$, $T_x/T_l$, and $T_x/T_g$, contribute evenly to the G-FAS criterion. By introducing an index $a$, the value of $(T_x/T_g)^a$=(1.0–1.3)±0.1 ($a$≈1.5±0.2) is close to the $T_g/T_l+T_x/T_l$= 0.49(Ni)~(0.74~1.48)~1.59($SiO_2$) value. Thus, the revised G-FAS criterion is obtained, as follows:

$$\text{G-FAS}_m'=T_g/T_l+T_x/T_l+(T_x/T_g)^{1.5} \quad (10)$$

## 3 Results and Discussion

### 3.1 Effectiveness of G-FAS criterion

In order to evaluate the effectiveness of the G-FAS criterion, a reliable database composed of a wide range of metallic glasses is chosen[44] for analysis. The optimal glass former $SiO_2$ and the worst glass former Ni are also included. The relationship between G-FAS criterion and lg$R_c$ is shown in Fig. 3, where an excellent linear relation can be clearly observed. The linear regression analysis result shows that the relationship between lg$R_c$ with G-FAS value can be expressed as follows:

$$\lg R_c=28.82-11.80\text{G-FAS} \quad (11)$$

where $R_c$ has the unit of K/s and G-FAS value is dimensionless. From the regression analysis of the plot between G-FAS criterion and lg$R_c$, the coefficient of determination $R^2$ is evaluated and recorded. The $R^2$ value can reflect the effectiveness and consistency of G-FAS criterion. The higher the $R^2$ value, the better the correlation between the proposed G-FAS criterion and $R_c$. In this case, the $R^2$ value is 0.882, suggesting that there is a solid correlation between $R_c$ and G-FAS criterion. The 95% prediction limits are also shown in Fig.3, as indicated by the dashed lines. A narrow band can be observed, inferring the less scatter of the experiment data and a stronger relationship between independent variables.

The G-FAS criterion, as an effective criterion for GFA&GS evaluation, can also be applied in other glass forming materials. Fig. 4 shows the correlations between the G-FAS criterion and critical cooling rate lg$R_c$ of 23 glassy oxides and 31 cryoprotective solutions[45]. The linear relationship can also be observed in Fig.4a and 4b, as demonstrated by solid lines. The relationship between the G-FAS criterion and critical cooling rate lg$R_c$ of 23 oxides can be expressed, as follows:

$$\lg R_c=31.94-13.30\text{G-FAS} \quad (12)$$

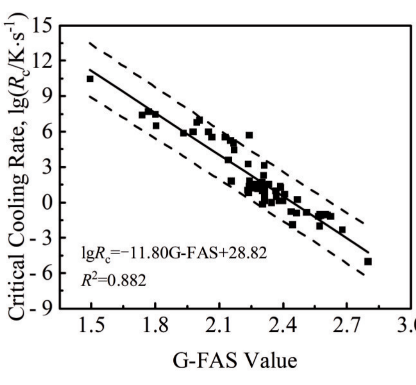


Fig.3 Correlation between G-FAS criterion and critical cooling rate lg$R_c$ of various metallic glasses

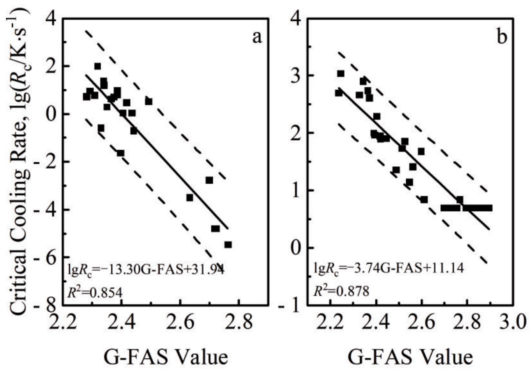


Fig.4 Correlations between G-FAS criterion and critical cooling rate lg$R_c$ of 23 glassy oxides (a) and 31 cryoprotective solutions (b)

The relationship between the G-FAS criterion and critical cooling rate lg$R_c$ of 31 cryoprotective solutions can be expressed, as follows:

$$\lg R_c=11.14-3.74\text{G-FAS} \quad (13)$$

The $R^2$ values of Eq.(12) and Eq.(13) are 0.854 and 0.878, respectively, suggesting that there is a relatively solid correlation between G-FAS criterion and critical cooling rate $R_c$.

It can be seen that the G-FAS criterion can be applied not only for metallic glasses, but also for the glass formers, presenting great application potential. Besides, this result also indicates that the G-FAS criterion for GFA&GS evaluation obtained by comprehensive consideration of GFA in cooling process and GS in heating process based on pseudo-four characteristic parameters is reliable.

According to the pseudo-four characteristic parameters and the derivation of the G-FAS criterion, it is verified that G-FAS criterion can simultaneously evaluate GFA and GS. Additionally, the characterization of correlation between proposed G-FAS criterion and GS-related parameter is necessary. It is well known that $T_x$ and $T_x-T_g$ can reflect GS. But for different glass formers, there is no contrast. Thus, considering the reduction of these two parameters with $T_g$, a quantitative representation of GS, the crystallization-vitrification ratio of $T_x/T_g$, is obtained for analysis. Based on the Tri-FAS, the ratio of $T_x/T_g$ can be used to reflect GS. Fig.5 shows the correlations between the modified G-FAS$_m$/G-FAS$_m'$ criteria and critical cooling rate lg$R_c$ and those between the modified G-FAS$_m$/G-FAS$_m'$ criteria and $T_{xg}$ for various metallic glasses. For the relationship between the modified criteria and $R_c$, the linear regression expressions are lg$R_c$=51.76−21.44× G-FAS$_m$ with $R^2$=0.895 and lg$R_c$=26.70−10.64G-FAS$_m'$ with $R^2$ =0.903 for the modified G-FAS$_m$ and G-FAS$_m'$ criteria, respectively. Compared with the G-FAS criterion ($R^2$=0.882), the modified criteria show stronger relationship with $R_c$ and higher $R^2$ values, indicating that the amelioration based on the competitive relationship between vitrification and crystallization during cooling process and the contribution of each component to the criterion is feasible. For the

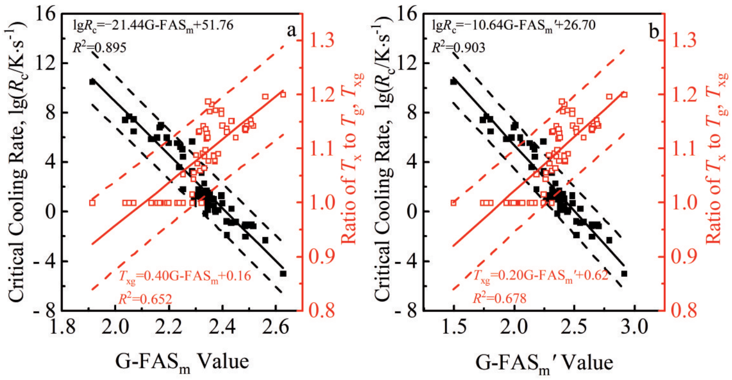


Fig.5 Correlations of modified G-FAS$_m$ criterion (a) and G-FAS$_m$′ criterion (b) with critical cooling rate $R_c$ and the ratio of $T_x/T_g$ ($T_{xg}$) for various metallic glasses

correlations between modified G-FAS criteria and $T_{xg}$, linear relationships between $T_{xg}$ and G-FAS$_m$/G-FAS$_m$′ criterion can be observed, and they can be expressed as follows:

$$T_{xg}=0.16+0.40\text{G-FAS}_m \quad (14)$$

$$T_{xg}=0.62+0.20\text{G-FAS}_m' \quad (15)$$

The coefficients of determination $R^2$ of the correlation between G-FAS$_m$ criterion and $T_{xg}$ and that between G-FAS$_m$′ criterion and $T_{xg}$ are 0.652 and 0.678, respectively. Compared with those from the correlation between lg$R_c$ and G-FAS$_m$/G-FAS$_m$′ criterion, the data from the correlation between $T_{xg}$ and G-FAS$_m$/G-FAS$_m$′ criterion are more scattered. This is probably attributed to the selection of GS parameter. However, the optimal parameter to evaluate GS is $T_{xg}$ in this research. In fact, similar to the critical cooling rate $R_c$ during cooling process, the critical heating rate $R_{hc}$ can also be used to reflect the crystallization resistance during heating process, i.e., $R_{hc}$ can reflect GA. However, considering the difficulty in measurement and the lack of available data of $R_{hc}$, $R_{hc}$ cannot be used as the GS parameter.

### 3.2 Comparison of relevant criteria

The items of G-FAS criterion were analyzed. It is worth noting that $T_{xg}$ is an indicator to determine the quality of other GS parameters. So $T_{xg}$ is not considered as a comparison item in this research. Table 1 shows the comparison results of determination coefficients $R^2$ of criterion components, $\gamma$-series criteria ($\gamma$, $\gamma_m$, $\gamma_c$, and $\gamma_n$), G-FAS, G-FAS$_m$, and G-FAS$_m$′ criteria for 65 metallic glasses, 23 glassy oxides, and 31 cryoprotective solutions. Firstly, from the perspective of GFA, the $T_{xl}$ term can simultaneously reflect GFA and GS and it has a larger $R^2$ value than the term $T_{rg}$ does, which only describes GFA, indicating that the $T_{xl}$ term is comparable to the G-FAS criterion in this research. However, the revised parameters of G-FAS$_m$ and G-FAS$_m$′ criteria are better than $T_{xl}$ term, especially for the metallic glasses. Secondly, from the perspective of GS, the $R^2$ values of G-FAS series criteria are higher than those of $T_{xl}$ and $T_{rg}$, indicating that G-FAS series criteria have stronger correlations with $R_c$, compared with the $T_{xl}$ and $T_{rg}$ terms.

It can be concluded that the parameters $T_{xl}$ and $T_{rg}$ cannot comprehensively evaluate GFA and GS. The $T_{xl}$ term considers the liquid stability and crystallization resistance, but ignores the GFA/glass-forming tendency and the SLR$_{glass}$ stability. The parameter $T_{rg}$ only considers the GFA reflected by the GFA/glass-forming tendency, but neglects the GFA&GS reflected by the liquid stability, crystallization resistance, and the SLR$_{glass}$ stability. Therefore, $T_{xl}$ and $T_{rg}$ are not the optimal criteria.

Finally, the G-FAS criteria with the commonly used $\gamma$-series criteria were discussed. Through the regression analysis, according to the relationships between lg$R_c$ and $\gamma$ criterion and those between $T_{xg}$ and $\gamma$ criterion for 65 metallic glasses, the $R^2$ values are 0.882 and 0.596, respectively, which are nearly equal to that of G-FAS criterion. This result indicates that G-FAS and $\gamma$ criteria are comparable in GFA&GS prediction. Correspondingly, the modified $\gamma$ criteria ($\gamma_m$, $\gamma_c$, and $\gamma_n$) and the revised G-FAS criteria (G-FAS$_m$ and G-FAS$_m$′) show comparable results in GFA&GS evaluation. In addition, from the expression of G-FAS criterion, it can be seen that three characteristic temperatures $T_x$, $T_l$, and $T_g$ exhibit the same times (2!). Similarly, in the $\gamma$ criterion, each characteristic

**Table 1 Comparison results of determination coefficients $R^2$ of criterion components, $\gamma$-series criteria ($\gamma$, $\gamma_m$, $\gamma_c$, and $\gamma_n$), G-FAS, G-FAS$_m$, and G-FAS$_m$′ criteria for various metallic glasses, glassy oxides, and cryoprotective solutions**

| Determination coefficient | Material | $T_{rg}$ | $T_{xl}$ | $\gamma$ | $\gamma_m$ | $\gamma_c$ | $\gamma_n$ | G-FAS | G-FAS$_m$ | G-FAS$_m$′ |
|---|---|---|---|---|---|---|---|---|---|---|
| $R^2$(GFA) | 65 metallic glasses | 0.646 | 0.846 | 0.882 | 0.901 | 0.908 | 0.908 | 0.882 | 0.895 | 0.903 |
| | 31 cryoprotective solutions | 0.547 | 0.854 | 0.879 | 0.868 | 0.858 | 0.864 | 0.878 | 0.874 | 0.879 |
| $R^2$(GS) | 23 glassy oxides | 0.809 | 0.889 | 0.787 | 0.771 | 0.575 | 0.673 | 0.854 | 0.761 | 0.736 |
| | 65 metallic glasses | 0.224 | 0.504 | 0.596 | 0.669 | 0.761 | 0.722 | 0.594 | 0.653 | 0.678 |

temperature also shows the same time (1!). This result indicates that the contribution of each characteristic temperature is even without bias. It should be noted that during the derivation process of $\gamma$ criterion, two aspects of $T_x/T_g$ and $T_x/T_l$[15] are considered, but the GFA/glass-forming tendency reflected by $T_g/T_l$ during cooling process is neglected. Although the modified $\gamma_m$, $\gamma_c$, and $\gamma_n$ criteria have a relatively high correlation with the critical cooling rate $R_c$[22,29,34], they probably cannot comprehensively and completely reflect GFA&GS, especially the GFA reflected by $T_g/T_l$ during the cooling process, under the consideration of the pseudo-four characteristic parameters obtained from both the cooling and heating processes.

## 4 Conclusions

1) Based on the four aspects reflected by the pseudo-four characteristic parameters, a simple triangle criterion for glass-forming ability (GFA) and the glass stability (GS) evaluations with the combination of pseudo-four characteristic parameters as the vertex is established under the consideration of both GFA during cooling process and GS during heating process, namely Tri-FAS.

2) Based on Tri-FAS and pseudo-four characteristic parameters, the G-FAS criterion is proposed and modified from the aspects of the competitive correlation between amorphization and crystallization during cooling process and the contribution of each component to the criterion. G-FAS criterion is as follows: G-FAS=$T_g/T_l+T_x/T_l+T_x/T_g$, G-FAS$_m$=$T_g/(1.5T_x)+T_x/T_l+T_x/T_g$, and G-FAS$_m$'=$T_g/T_l+T_x/T_l+(T_x/T_g)^a$ ($a\approx1.5\pm0.2$), where $T_g$ is glass transition temperature, $T_x$ is onset crystallization temperature, and $T_l$ is liquid temperature.

3) For various metallic glasses and glass formers, the G-FAS criterion exhibits the better correlation than $T_g/T_l$ and $T_x/T_l$ items do for the GFA&GS evaluation. G-FAS criterion is comparable to $\gamma$ criterion for GFA&GS evaluation, which is feasible and reliable.

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# 玻璃形成能力及稳定性三角形判定准则


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**摘 要：**根据降温过程的玻璃形成能力（GFA）和升温过程的玻璃稳定性（GS），构建了以伪四特征参数组合为顶点的判定玻璃形成能力和稳定性的三角形（Tri-FAS），从而推导出判定GFA&GS的准则：G-FAS=$T_g/T_l+T_x/T_l+T_x/T_g$（$T_x$为起始结晶温度；$T_l$为液体温度；$T_g$为玻璃化转变温度），并从降温过程非晶化与晶化之间的竞争关系和准则各组成项对准则的均衡贡献两个方面进行了修订：G-FAS$_m$=$T_g/(1.5T_x)+T_x/T_l+T_x/T_g$和G-FAS$_m'$=$T_g/T_l+T_x/T_l+(T_x/T_g)^a$ ($a$≈1.5±0.2)。讨论了G-FAS与临界冷却速率$R_c$、G-FAS与$T_{xg}$（$T_{xg}$反映了玻璃的过冷液区，$T_{xg}=T_x/T_g$）的相关性，分别能够反映GFA和GS。通过大量金属玻璃和其他玻璃形成体从GFA和GS两方面对判定准则的有效性进行了评估，结果显示：该判定准则无论是GFA方面还是GS方面，在不同玻璃形成体系中均可靠有效，具有广泛应用性。提出的Tri-FAS和G-FAS判定准则在玻璃的生产和实际应用过程中具有指导作用。



**关键词：**金属玻璃；玻璃形成能力；热稳定性；特征温度；准则



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