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Correlation between fragility and glass-forming ability of metallic alloys

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Analysis of the relaxation time of the glass forming liquids at near liquidus temperatures was conducted, correlation between the critical cooling rate for glass formation, fragility of the glass forming liquid, and reduced glass transition temperature was identified, and a glass forming ability (GFA) parameter ($F_1 = 2[(m/m_{\min})(1/T_{rg} - 1) + 2]^{-1}$) was proposed. This GFA parameter, which increases with a decrease in the critical cooling rate, is a function of the reduced glass transition temperature T_{rg} and a fragility index m, and it varies from ~ 0 in the case of extremely fragile liquid to $2T_{rg}/(1+T_{rg})$ in the case of extremely strong liquid. An exponential relationship between the critical cooling rate for glass formation and the parameter F_1 was identified and verified using available experimental data for metallic and nonmetallic glasses.

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INTRODUCTION

Over the last two decades, a number of bulk metallic glasses have been produced with the thicknesses from 1 mm to several centimeters, and empirical methods have been proposed[1,2] to guide the discovery of new glasses with better glass forming ability (GFA). A straightforward indicator of GFA is the critical cooling rate R_c, above which no crystallization occurs during solidification, and lower R_c always correspond to higher GFA.[2,3] Although this parameter allows a way to directly validate and compare GFA of different alloys, a number of solidification trials with varying cooling rates of cast material are required to determine R_c, which is a time and labor consuming process. Thus, other parameters, which can easily be determined from differential thermal analysis, have been proposed and tried to correlate with R_c.[2,4–6] The most extensively used empirical parameters are the reduced glass transition temperature, $T_{rg} = T_g/T_l$, a supercooled liquid range, $\Delta T_x = T_x - T_g$, and the parameter $\gamma = T_x/(T_g + T_l)$. Here, T_g, T_x, and T_l are the glass transition, crystallization, and liquidus temperatures, respectively. Although each of these empirical relations can correlate quite well with GFA of some alloys, the correlation can be poor for other alloy systems.[4–6] There is also not yet a clear answer on what physical factors these relations are based.

It has recently been perceived that good glass forming alloys generally have strong liquids with high viscosity, while marginal glass formers have more fragile liquids.[7–9] Though no direct correlation between GFA and fragility has yet been identified,[10,11] it was suggested that GFA can be a complex function of fragility and T_{rg} (Ref. 10) or fragility and the onset driving force for crystallization.[12] This paper is focused on identifying a fundamental relationship between fragility and GFA in metallic and nonmetallic glass formers.

FRAGILE BEHAVIOR OF SUPERCOOLED LIQUID

During supercooling of liquid, the relaxation time (τ) for atom rearrangements can increase in a non-Arrhenius fashion by many orders of magnitude in a narrow temperature range. The magnitude of departure from Arrhenius behavior is called liquid fragility[13,14] and the non-Arrhenius behavior of $\tau(T)$ can be described by the Vogel-Fulcher-Tammann (VFT) expression[13]

$$\tau = \tau_o \exp\left(\frac{DT_o}{T - T_o}\right), \tag{1}$$

where τ_o, D, and T_o are parameters and T is the temperature in Kelvin. At $D \geqslant 100$, the behavior is very much like Arrhenius and with D decreasing to below 10, the behavior becomes more fragile. The parameter D is therefore called the strength parameter.[14] The parameter T_o depends on D in such a way that when D approaches infinity, T_o moves toward zero and when D approaches zero, T_o becomes equal to T_g. Indeed, at $T = T_g$, the logarithmic form of Eq. (1) reads

$$\log_{10}\left(\frac{\tau_g}{\tau_o}\right) = \frac{DT_o}{(T_g - T_o)\ln 10}, \tag{2}$$

where τ_g is the relaxation time at T_g. By definition, $\tau_g = 100$ s,[13] while experimental data indicate that for many liquids, $\tau_o \approx 10^{-14}$ s.[14–17] This leads to the following relationship between D and T_o:

$$D = \frac{m_{\min}(T_g - T_o)}{T_o} \ln 10, \tag{3}$$

where $m_{min} \equiv \log_{10}(\tau_g/\tau_o) \approx 16$. By combining Eqs. (1) and (3), we obtain a modified VFT relation,

$$\log_{10}\left(\frac{\tau}{\tau_o}\right) = m_{\min}\frac{T_g - T_o}{T - T_o}. \tag{4}$$

A convenient method to measure fragility of glass forming liquids has recently been suggested[14–16] by using a fragility index m,

$$m = \frac{d \log_{10} \tau}{d(T_g/T)_{T=T_g}}. \tag{5}$$

Liquids with $m \approx m_{\min}$ show the Arrhenius behavior, and larger departure of m from $m_{\min}$ indicates higher fragility. A relation between m and T_o is obtained by differentiating Eq. (4),

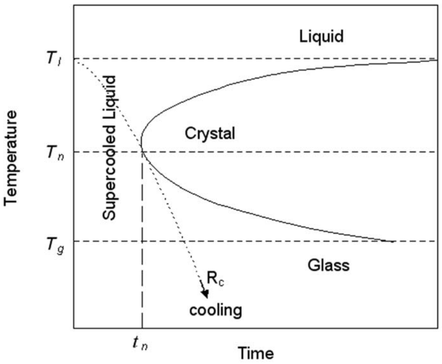

FIG. 1. Time-temperature transformation diagram for the onset of crystallization of a glass forming liquid (schematic). Crystallization occurs between T_l and T_g, and it can be avoided when the liquid is chilled with the cooling rate $R \geqslant R_c$.

$$\frac{1}{m} = \frac{1}{m_{\min}}\left(1 - \frac{T_o}{T_g}\right). \tag{6}$$

A relation between m and D can be obtained by combining Eqs. (3) and (6),

$$D = \frac{m_{\min}^2}{m - m_{\min}} \ln 10. \tag{7}$$

CORRELATION BETWEEN LIQUID FRAGILITY AND THE CRITICAL COOLING RATE FOR GLASS FORMATION

We will now identify the correlation between the liquid fragility and glass forming ability. Liquid can be undercooled and form the glassy state only if crystallization is kinetically constrained due to a rapid increase in the viscosity and relaxation time during quenching. Crystallization may occur in the temperature interval between T_l and T_g and an incubation time is required to start the crystallization.[4,18] The temperature dependence of the incubation time [a so-called time-temperature transformation (TTT) diagram, Fig. 1] has a C shape, and the minimum time t_n, which is required to start crystallization, occurs at a temperature T_n located between T_g and T_l. This temperature can be expressed in terms of T_g and T_l as $T_n = a(T_g + T_l)$, where parameter a is between 0.45 and 0.55, and it is generally close to 0.5.[18–20] To avoid crystallization, the cooling curve should not intersect the TTT curve, and the minimum (critical) cooling rate R_c required to form a glass is the cooling rate needed to bypass the nose of the TTT curve (see Fig. 1).

Therefore, R_c is inversely proportional to t_n and can be virtually defined as $R_c \approx (T_l - T_n)/t_n$. It is reasonably to assume that t_n is proportional to the relaxation time τ_n of the supercooled liquid at $T = T_n$, which can be determined from Eq. (4) as

$$\log_{10}\left(\frac{\tau_n}{\tau_o}\right) = \frac{m_{\min}(T_g - T_o)}{T_n - T_o} \approx \frac{m_{\min}(T_g - T_o)}{0.5(T_l + T_g) - T_o}. \tag{8}$$

Therefore, $\log_{10}(R_c) \sim \log_{10}(1/\tau_n) = -m_{\min}(T_g - T_o)/[0.5(T_l + T_g) - T_o]$. Because the glass forming ability increases with a decrease in R_c, a GFA parameter F_1, which is proportional to $-\log_{10}(R_c)$, can be defined,

$$F_1 = \frac{(T_g - T_o)}{0.5(T_l + T_g) - T_o} \sim -\log_{10}(R_c). \tag{9}$$

According to Eq. (9), F_1 increases and R_c decreases with an increase in T_g and a decrease in T_o and T_l. In the extreme case of very strong liquid ($T_o \approx 0$), $F_1 = 2T_{rg}/(1 + T_{rg})$, where $T_{rg} = T_g/T_l$. For very fragile liquids, when T_o approaches T_g, F_1 will be close to zero.

Taking into account the relation between T_o and D [Eq. (3)] or between T_o and m [Eq. (6)], F_1 can also be expressed in terms of T_{rg} and D or T_{rg} and m,

$$F_1 = \frac{2T_{rg}D}{D(1 + T_{rg}) + m_{\min}(1 - T_{rg})\ln 10} \tag{10}$$

or

$$F_1 = 2\left[\frac{m}{m_{\min}}\left(\frac{1}{T_{rg}} - 1\right) + 2\right]^{-1}. \tag{11}$$

The data presented in Tables I and II were used to validate the GFA parameter F_1 and identify the correlation between F_1 and R_c. To illustrate generality of the present analysis, not only a broad range of metallic glasses but also several nonmetallic glasses were used for which R_c values are known and other parameters, which are required to determine F_1, are also available experimentally. Table I lists glasses for which reliable parameters T_o and D are available, and m was calculated using $m = DT_oT_g/(T_g - T_o)^2 \ln 10$.[15] Two values of the parameter F_1, $F_1(T_o)$ and $F_1(D)$, listed in this table for each alloy, were calculated using Eqs. (9) and (10), respectively, and their average values were used for validation.

Table II lists the glasses for which reliable m values are available or they can be determined using Eq. (5) from the experimentally accessible temperature dependences of the relaxation time or viscosity near T_g. Correspondingly, Eq. (11) was used to calculate F_1 listed in this table. Figure 2 shows correlation between R_c and the GFA parameter F_1. The best linear fit of these experimental data points in the coordinates $\log_{10}(R_c)$ vs F_1 was conducted using the least squares method regression analysis, and the following relation was identified:

$$R_c = R_{co} \exp(-AF_1), \tag{12}$$

where $R_{co} \approx 2.7 \times 10^{11}$ K/s and $A \approx 48.7$. A rather high value of the correlation coefficient (or the coefficient of determination, $R^2 = 0.962$) indicates that Eq. (12) fits the experimental points very well. If relation (12) is valid within a whole range of F_1, then it predicts R_c maximum for the extremely fragile liquid ($F_1 = 0$), $(R_c)_{\max} \equiv R_{co} \approx 2.7 \times 10^{11}$ K/s. This value is in good agreement with the critical cooling rate range for pure metals.[2,4] For the extremely strong liquid, when $T_{rg} \approx 2/3$ (Ref. 21) and $F_1 \approx 0.8$, Eq. (12) predicts the minimum critical cooling rate $(R_c)_{\min} \approx 2.3 \times 10^{-6}$ K/s, which can be compared with $R_c \approx 10^{-5}$ K/s for SiO_2.[31,32]

TABLE I. The critical cooling rate R_c, characteristic temperatures (T_g, T_l, T_o), reduced glass transition temperature T_{rg}, strength parameter D, fragility index m, and GFA parameter F_1 for several bulk metallic glasses and two oxide glasses.

Glass formers	R_c (K/s)	T_g (K)	T_l (K)	T_o (K)	D	m	F_1 (D)	F_1 (T_o)
$Mg_{65}Cu_{25}Y_{10}$[a]	50	402	730	260	22.1	50	0.479	0.464
$Pd_{40}Ni_{40}P_{20}$[b]	1.6	578	973	390	18.1	50	0.491	0.488
$Pd_{48}Ni_{32}P_{20}$[c]	10	590	985	392	16.6	43	0.481	0.501
$Pd_{77.5}Cu_6Si_{16.5}$[d]	500	637	1058	493	11.1	73	0.412	0.406
$Cu_{47}Ti_{34}Zr_{11}Ni_8$[e]	250	673	1128	500	12.0	59	0.421	0.432
$Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$[f]	1.2	625	993	412.5	18.5	46	0.532	0.536
$Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$[g]	8.0	590	1020	372	22.7	46	0.511	0.503
SiO_2[h]	10^{-5}	1452	1999	530	63.3	25	0.770	0.771
GeO_2[h]	10^{-3}	816	1388	199	112.6	21	0.683	0.683

[a] Reference 22.
[b] References 23 and 24.
[c] Reference 25.
[d] References 25 and 26.
[e] References 7 and 25.
[f] References 27–29.
[g] References 25 and 30.
[h] References 31 and 32.

TABLE II. The critical cooling rate R_c, glass transition T_g, and liquidus T_l temperatures, fragility index m, and GFA parameter F_1 for several bulk metallic glasses and nonmetallic glasses.

Glass formers	R_c (K/s)	T_g (K)	T_l (K)	m	F_1
$La_{55}Al_{25}Ni_{20}$[a]	89	491	941	42	0.455
$La_{55}Al_{25}Cu_{20}$[b]	109	456	896	43	0.435
$La_{55}Al_{25}Ni_{15}Cu_5$[c]	20	472	900	37	0.485
$La_{55}Al_{25}Ni_{10}Cu_{10}$[c]	10	467	835	35	0.540
$La_{55}Al_{25}Ni_5Cu_{15}$[c]	43	459	878	42	0.453
$La_{55}Al_{25}Ni_5Cu_{10}Co_5$[c]	7.9	466	823	37	0.530
$Pd_{40}Ni_{10}Cu_{30}P_{20}$[d]	0.1	577	836	52	0.578
$Zr_{65}Al_{7.5}Ni_{10}Cu_{17.5}$[e]	1.5	656	1168	35	0.539
Silica[f]	10^{-4}	1393	2003	28	0.687
Germania[g]	10^{-2}	820	1383	24	0.660
$CaO \cdot MgO \cdot 2SiO_2$[h]	169	995	1664	66	0.419
$Na_2O \cdot 2SiO_2$[i]	1.7×10^{-3}	708	1135	29	0.647
$Li_2O \cdot 2SiO_2$[i]	1.0	710	1316	33	0.532
Salol[j]	1.0	218	313	66	0.521

[a] References 33–35.
[b] References 33 and 34.
[c] References 34 and 35.
[d] References 6, 8, and 36.
[e] References 6, 8, and 37.
[f] References 14, 38, and 39.
[g] References 39 and 40.
[h] References 41 and 42.
[i] Reference 43.
[j] References 31, 40, and 44.

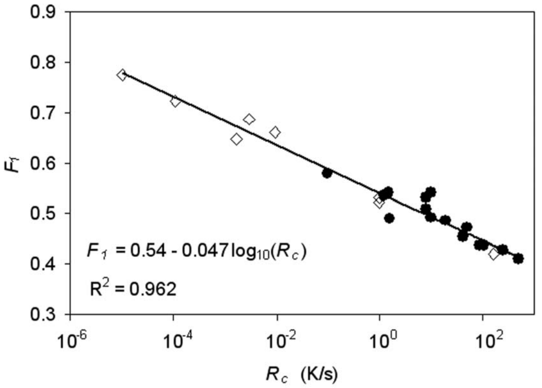

FIG. 2. Correlation between the critical cooling rate R_c and the glass forming ability parameter F_1 for several glass forming systems listed in Tables I and II. Solid circles correspond to the bulk metallic glasses and open diamonds correspond to the nonmetallic glasses.

Figure 3(a) shows correlation between R_c and T_{rg} for the glass formers from Tables I and II. Although there is a trend for R_c to decrease with an increase in T_{rg}, the correlation is not very good ($R^2=0.451$). This indicates that T_{rg} cannot be a reliable GFA parameter. Similar conclusion regarding T_{rg} was also made in other publications.[6,10,12] In accord to Eqs. (9)–(11), T_{rg} can be used as a GFA indicator only for glass forming systems in which fragility weakly depends on the composition.

Figure 3(b) shows dependence of R_c on the fragility index m. There is a trend for R_c to decrease with a decrease in m. However, for metallic glasses, a "correct" tendency for R_c to decrease with a decrease in m is observed only at R_c of $\sim$10 K/s and higher. At the lower R_c, a reverse correlation can be noticed, when R_c decreases with an increase in m. Overall, the correlation between these two parameters is not very good, and the correlation coefficient is low, $R^2=0.454$. No correlation between R_c and fragility for bulk metallic glasses was also reported in other works,[10–12,45] although some tendency for R_c to increase with an increase in the fragility index was noticed for the glass forming alloys with $R_c > 10$ K/s.[45]

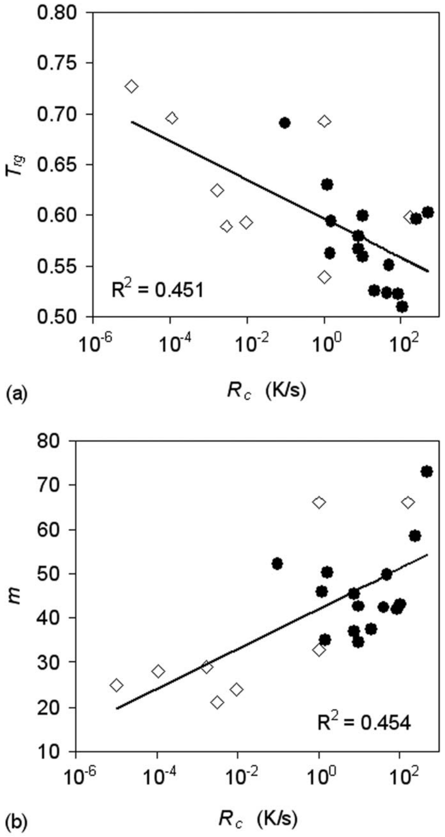

FIG. 3. Dependence of the critical cooling rate R_c on (a) the reduced glass transition temperature $T_{rg}=T_g/T_l$ and (b) the fragility parameter m for several glass formers listed in Tables I and II. Solid circles correspond to the bulk metallic glasses and open diamonds correspond to the nonmetallic glasses.

DISCUSSION

Many researchers use T_{rg} or m to estimate GFA of different systems.[3–6] These parameters, when used independently, correlate well with the GFA (i.e., with R_c) of some glass forming systems but the correlations are poor for other glass forming systems,[6,9–12,45] and Fig. 3 is an example. Until now, there was no clear explanation of such unpredictable behavior. The analysis conducted in the present work explains the reason of such inconsistency. Indeed, this analysis shows that R_c is a function of both T_{rg} and m and, therefore, these parameters must be used together to estimate GFA. The parameter T_{rg} alone can be a good GFA indicator only for glass forming systems which have similar fragility values. However, T_{rg} cannot be a reliable GFA parameter for materials with significantly different fragilities [see Fig. 3(a)]. Similarly, good correlation between GFA and liquid fragility can be observed only for the systems which have nearly the same T_{rg} values. In general, however, both the reduced glass transition temperature and the fragility must be known in order to predict GFA (R_c) of a liquid. As a result, a GFA parameter F_1, which is proportional to $-\log_{10}(R_c)$ and is a function of T_{rg} and m, was introduced [see Eqs. (9)–(11)].

The physical background for such GFA behavior is illustrated in Figs. 4 and 5. Figure 4 illustrates the dependence of the GFA parameter F_1 on m for hypothetical liquids, which have three different T_{rg} values: 0.4, 0.55, and 0.67. Two horizontal dotted lines corresponding to $F_1=0.4$ ($R_c=940$ K/s) and $F_1=0.6$ ($R_c=0.06$ K/s) are also drawn, indicating that the liquids with a higher F_1 value have better GFA. These dotted lines cross the $F_1(m,T_{rg})$ lines at given values of m, which decrease with a decrease in T_{rg}. Taking into account that liquid fragility increases with an increase in m, one can see that liquids with higher T_{rg} must be more fragile in order to have the same R_c values. This figure illustrates that (a) each specific value of F_1 defines a particular critical cooling rate R_c, (b) liquids with different m values can have the same R_c, (c) liquids with different T_{rg} values can have the same R_c, and (d) liquids with higher fragility (higher m) must have higher T_{rg} values in order to have the same GFA (R_c). This

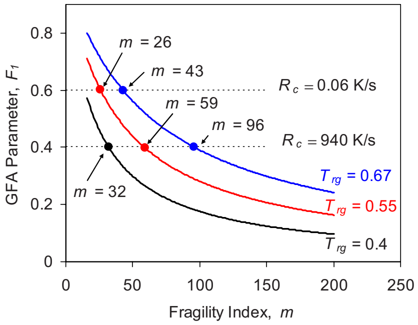

FIG. 4. (Color online) Dependence of the glass forming ability parameter F_1 [$=0.54-0.047\log_{10}(R_c)$] on the fragility index m at three different T_{rg} values.

clearly explains why, when they are used individually, the parameters T_{rg} and m cannot predict the glass forming ability. However, when these two parameters are combined into the GFA parameter F_1 through Eq. (11), they predict the glass forming ability of different families of glass forming liquids very well (see Fig. 2).

Figure 5 shows the temperature dependences of the relaxation time of seven glass forming liquids from Table I in the vicinity of the liquidus temperatures. It can be seen from this figure that the alloy $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ is the strongest and the alloy $Pd_{77.5}Cu_6Si_{16.5}$ is the most fragile among these seven alloys. At a given normalized reciprocal temperature (T_g/T), a stronger liquid has a longer relaxation time τ and, therefore, longer incubation period for starting crystallization. However, at the normalized temperature T_n [$T_g/T_n \cong 2T_{rg}/(1+T_{rg})$], which corresponds to the nose of the TTT crystallization curve (see Fig. 1) and which values are different for different alloys (they are shown as open points on the respective relaxation curves in Fig. 5), the straight dependence of the relaxation time τ_n (and, therefore, GFA, which is proportional to $\log_{10}\tau_n$) on liquid fragility may no longer be valid. A stronger liquid with a smaller T_{rg} may have a shorter τ_n and start to crystallize faster than a more fragile liquid with a higher T_{rg}. For example, the $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$ liquid is stronger than $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (compare, respectively, lines 7 and 5 in Fig. 5). However, the latter alloy has higher T_{rg} (=0.629 versus 0.578), which leads to a higher value of the relaxation time at T_g/T_n and, therefore, better GFA of the $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ alloy (see Fig. 5 and Table I). On the other hand, alloys $Pd_{77.5}Cu_6Si_{16.5}$, $Cu_{47}Ti_{34}Zr_{11}Ni_8$, $Pd_{40}Ni_{40}P_{20}$, and $Pd_{48}Ni_{32}P_{20}$ have almost the same T_{rg} (i.e., the same T_g/T_n) values, see Fig. 5. Therefore, for these four alloys, the GFA increases with a decrease in fragility.

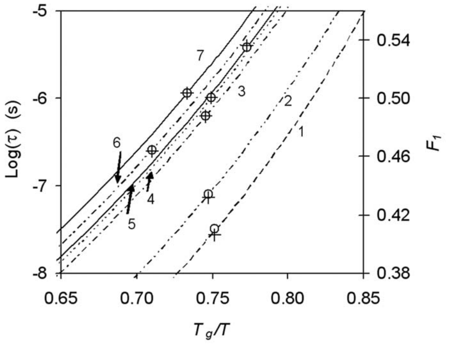

FIG. 5. Fragility plot of the relaxation times τ (lines) of seven glass forming alloys. The lines, in the order from the bottom to the top, correspond to (1) $Pd_{77.5}Cu_6Si_{16.5}$, (2) $Cu_{47}Ti_{34}Zr_{11}Ni_8$, (3) $Pd_{40}Ni_{40}P_{20}$, (4) $Pd_{48}Ni_{32}P_{20}$, (5) $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$, (6) $Mg_{65}Cu_{25}Y_{10}$, and (7) $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$. Open circles on the lines correspond to the nose temperatures T_n [i.e., $T_g/T_n \cong 2T_{rg}/(T_{rg}+1)$ in this plot] of the respective alloys. The F_1 values of these alloys are also plotted here (as crosses) versus respective T_g/T_n values.

CONCLUSIONS

Analysis of the fragile behavior and relaxation time of glass forming liquid in the temperature range between the liquidus temperature and glass transition temperature allowed us to identify unique correlation between the critical cooling rate for glass formation R_c, liquid fragility index m, and reduced glass transition temperature T_{rg}. Correspondingly, a GFA parameter F_1, which is proportional to $-\log_{10}(R_c)$ and is a function of T_{rg} and m, was proposed. This GFA parameter increases with an increase in T_{rg} and a decrease in m in accord to Eq. (11) and varies from ~ 0 in the case of the extremely fragile liquid (e.g., pure metals) to ~ 0.8 in the case of the extremely strong liquid (e.g., SiO_2). Validity and universality of this GFA parameter F_1 was experimentally verified for a number of bulk metallic and nonmetallic glasses. On the other hand, when used alone, T_{rg} and m were shown to be able to predict GFA only in special cases.

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