

Universal Scaling in the Temperature-Dependent Viscous Dynamics of Metallic Glasses

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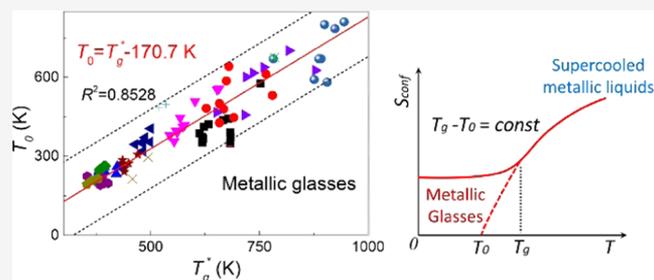
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ABSTRACT: The essential query about glass formation is how to understand the sheer temperature dependence of viscous dynamics of glass-forming liquids near the liquid-to-glass-transition temperature T_g . In this work, we report a universal scaling in the temperature-dependent viscous dynamics of metallic glasses (MGs) in the form of the Williams–Landel–Ferry equation on the basis of compiled data on the temperature-dependent viscosity and structural relaxation times of 89 MGs ever-reported in the past decades. Implications of this universal scaling are illustrated in the framework of the Adam–Gibbs relation, suggesting a universal vitrification mechanism in MGs mediated by configurational entropy wherein configurational entropy vanishes universally for all supercooled metallic liquids after a further decrease in temperature of ~ 170.7 K (whereas with a relatively large error of ± 150 K) below T_g . This result corroborates the thermodynamic origin of glass formation and suggests that MGs are an ideal research subject for understanding in depth the nature of glass formation for their relatively simple molecular structures.



1. INTRODUCTION

The central feature of the forming process of a glass is the drastically slowing-down viscous dynamics of the glass-forming liquid upon decreasing temperature.¹ While glass formation is omnipresent in diverse matters, e.g., polymers, small molecular liquids, oxides, metallic alloys, etc., and being investigated for decades, the physical mechanisms governing the temperature-dependent viscous dynamics remain unclear.² As early as in 1955, Williams et al.^{3,4} had observed an empirical universal formulation as eq 1, i.e., the Williams–Landel–Ferry (WLF) equation, for the temperature-dependent viscosity and electrical relaxation times of a variety of glass-forming liquids, such as polymers, molecular glasses, and oxide glasses, in the vicinity of a reference temperature $T_s = T_g + 50$ K, where T_g is the liquid-to-glass transition temperature; $\eta(T_s)$ is the viscosity at the reference temperature T_s ; and $c_1^s = 8.86$ and $c_2^s = 101.6$ K are constant coefficients for all of these glass-forming liquids.

$$\log \frac{\eta(T)}{\eta(T_s)} = \frac{-c_1^s(T - T_s)}{c_2^s + (T - T_s)} \quad (1)$$

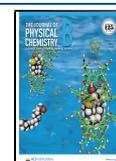
This universal formulation was first attributed to a universal temperature dependence of fractional free volume, and it was suggested that free volume determined the viscous dynamics of these glass systems.⁵ However, for the difficulty in defining free volume, especially in polymers due to their complicated molecular structures, Adam and Gibbs⁶ rationalized this universal formulation from the perspective of a thermodynamic property, i.e., configurational entropy, which refers to the

excess entropy in the equilibrium melt over the crystal. The universality in diverse glass-forming liquids was interpreted as a coincidence arising from a “semiuniversal” temperature dependence of configurational entropy near T_g , and it was concluded that configurational entropy governed glass formation independent of molecular structures. However, assessment on the contributions to excess entropy from configurational part and vibrational part in molecular and network glasses^{7–9} suggests that the excess entropy comprises both an anharmonic vibrational part and a configurational part. This result undermines the basis of the Adam–Gibbs relation that the excess entropy is mainly configurational and questions the crucial role of configurational entropy in glass formation. More recently, the vibrational entropy in metallic glasses (MGs) has been evaluated by in situ measurement of their vibrational spectra,¹⁰ exhibiting a tiny excess over their crystalline counterparts, which represents less than 5% of the total excess entropy. The results suggest that, in contrast to molecular and network glasses, the excess entropy in MGs is mainly composed of configurational entropy and that glass formation in MGs is accommodated solely by a change in

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configurational entropy. Thereby, it is expected that examining the validity of the universal WLF formulation in MGs, which has yet not been done, would effectively help in unveiling the role played by configurational entropy in the temperature-dependent viscous dynamics of glass-forming liquids.

In the following content, the measures to quantitatively describe the temperature dependence of viscous dynamics in glass-forming liquids, i.e., the fragility, were reintroduced in detail, and then how to examine the universal WLF formulation in MGs based on the fragility data was illustrated. Next, compiled data on the fragility of 89 MGs ever-reported in the past decades and their validity were analyzed. Based on these data, a universal scaling of WLF formulation was uncovered in the temperature dependence of viscous dynamics for MGs. This universal scaling in MGs was compared to the observations made by Williams et al.⁴ in the framework of free volume theory. Finally, this universal scaling was rationalized based on the Adam–Gibbs relation and unraveled a universal configurational entropy origin in the glass formation of MGs.

2. METHODS

First, we reviewed the concept of fragility in MGs and outlined the intercorrelations between the fragility parameters of MGs. Increasing evidences^{11–13} suggest that glass-forming systems generally exhibit a Vogel–Fulcher–Tammann (VFT)^{14–16}-type temperature dependence in viscosity as eq 2

$$\eta(T) = \eta_0 \cdot \exp\left(\frac{D \cdot T_0}{T - T_0}\right) \quad (2)$$

where η_0 is the high-temperature limit of viscosity, D is a material-dependent constant representing the deviation of the temperature dependence of viscosity from the Arrhenius relation, and T_0 is the ideal glass-transition temperature. To distinguish the temperature dependence of viscosity for different glass-forming liquids, the concept of “fragility” was proposed by Angell^{17,18} as the slope of the $\log \eta(T)$ vs $\frac{T_g^*}{T}$ plot

at the glass-transition temperature T_g^* , i.e., $m = \frac{d \log \eta}{d(T_g^*/T)}|_{T=T_g^*}$, where T_g^* is the temperature at which the viscosity of glass-forming liquids equals 10^{12} Pa·s or the relaxation time equals 100 s. T_g^* is close to the calorimetric glass-transition temperature T_g measured at a heating rate of 1 K/min.¹⁹ Since the heating rate dependence of T_g is insignificant,¹ here, T_g^* and T_g could be used interchangeably within experimental error as will be shown later. Based on eq 2, the fragility m reads

$$m = \frac{D \cdot (T_0/T_g^*)}{\left(1 - \frac{T_0}{T_g^*}\right)^2 \ln 10} \quad (3)$$

Recalling the definition of T_g^* : the temperature at which $\eta(T_g^*) = 10^{12}$ Pa·s, from eq 2, we have

$$\eta(T_g^*) = \eta_0 \cdot \exp\left(\frac{D \cdot T_0}{T_g^* - T_0}\right) \quad (4)$$

$$\frac{D \cdot T_0}{T_g^* - T_0} = 12 \ln 10 - \ln \eta_0 = \beta \quad (5)$$

$$T_g^*/T_0 = 1 + D/\beta \quad (6)$$

The value of η_0 could be estimated as: $\eta_0 = N_A h/V$,²⁰ where N_A is Avogadro's constant 6.022×10^{23} , h is Planck's constant 6.626×10^{-34} J·s, and V is the molar volume of the liquids, which is usually of an order of 10^{-5} m³. Thereby, Nemilov²¹ proposed that η_0 is a constant of 4×10^{-5} Pa·s for all liquids. Based on eq 5, β equals a constant 37.75 and suggests a linear correlation between D and (T_g^*/T_0) (see eq 6). This constant β has been reported as ~ 38 in the $\log \eta$ vs $\frac{T_g - T_0}{T - T_0}$ plot proposed by Takeuchi et al.²² It can be seen that β is determined by the value of η_0 and the definition of T_g^* . By substituting eq 6 into 3, the relationship between D and m reads

$$m = \frac{\beta}{\ln 10} + \frac{\beta^2}{\ln 10} / D \quad (7)$$

The relationship between m and D has been proposed previously as: $m = 16 + 590/D$.¹⁸ Based on eqs 5 and 7, it is seen that this equality was derived by assuming $\eta_0 = 10^{-4}$ Pa·s, which corresponds to $\beta = 36.84$. This fact indicates that the relationship between m and D is affected by the value of η_0 . Equation 7 also indicates that the fragility m could be uniquely determined from the value of D . Namely, D is equivalent to m in measuring the fragility of glass-forming liquids. In the strong extreme, with D approaching infinity or $m \approx 16$,¹⁸ the liquid exhibits an Arrhenius-type temperature dependence in viscosity, while in the fragile extreme, with $D \approx 3.8$ or $m \approx 170$,²³ the liquid exhibits a super-Arrhenius-type behavior in viscosity.²⁴

Second, we illustrated how to examine the universal WLF formulation in MGs based on the fragility data. Based on an arbitrarily chosen reference temperature T_{ref} , the empirical WLF equation, i.e., eq 1, could be written in a general form as⁶

$$\log \frac{\eta(T)}{\eta(T_{\text{ref}})} = \frac{-C_1^{\text{ref}}(T - T_{\text{ref}})}{C_2^{\text{ref}} + (T - T_{\text{ref}})} \quad (8)$$

In eq 8, c_1^{ref} and c_2^{ref} are coefficients associated with the chosen value of T_{ref} . The formulation reported by Williams et al.⁴ is actually the case in which $T_{\text{ref}} = T_s = T_g + 50$ K, $c_1^s = 8.86$, and $c_2^s = 101.6$ K. For c_1^s and c_2^s , the superscript s represents the subscript of the reference temperature T_s , since the reference temperature could be arbitrarily chosen.

The relationship between $c_1^s = 8.86$, $c_2^s = 101.6$ K and c_1^{ref} , c_2^{ref} could be derived from⁶

$$\log \frac{\eta(T)}{\eta(T_{\text{ref}})} = \log \frac{\eta(T)}{\eta(T_s)} - \log \frac{\eta(T_{\text{ref}})}{\eta(T_s)} \quad (9)$$

With proper reorganization, the above expression gives

$$c_1^{\text{ref}} = c_1^s c_2^s / (c_2^s - (T_s - T_{\text{ref}})); \quad c_2^{\text{ref}} = c_2^s - (T_s - T_{\text{ref}}) \quad (10)$$

Based on these formulas, Williams et al.⁴ provided $c_1^g = 17.44$ and $c_2^g = 51.6$ K when T_g was chosen as the reference temperature. The superscript g represents the subscript of the reference temperature T_g . However, as will be shown later, the physical meanings of the two coefficients are not explicit.

Furthermore, the formulation of the WLF equation could be readily derived from the VFT equation. Introducing eq 2 on the left-hand side of eq 8, we have

$$\begin{aligned} \log \frac{\eta(T)}{\eta(T_{\text{ref}})} &= \left(\frac{D \cdot T_0}{T - T_0} - \frac{D \cdot T_0}{T_{\text{ref}} - T_0} \right) \cdot \frac{1}{\ln 10} \\ &= \frac{D \cdot T_0 / \ln 10 \cdot (T_{\text{ref}} - T)}{(T - T_0)(T_{\text{ref}} - T_0)} \\ &= \frac{-D \cdot T_0 / \ln 10 \cdot (T_{\text{ref}} - T_0) \cdot (T - T_{\text{ref}})}{(T_{\text{ref}} - T_0) + (T - T_{\text{ref}})} \end{aligned} \quad (11)$$

Comparing the last terms in eqs 11 with 8, we have

$$c_1^{\text{ref}} = D \cdot T_0 / \ln 10 (T_{\text{ref}} - T_0) \quad (12)$$

$$c_2^{\text{ref}} = T_{\text{ref}} - T_0 \quad (13)$$

By choosing $T_{\text{ref}} = T_g^*$, the above expressions become: $c_1^{\text{ref}} = D \cdot T_0 / \ln 10 (T_g^* - T_0)$ and $c_2^{\text{ref}} = T_g^* - T_0$. From eq 5, $\beta = \frac{D \cdot T_0}{T_g^* - T_0}$ equals a constant 37.75. Thereby, $c_1^{\text{ref}} = \beta / \ln 10$ is also a constant 16.4, indicating that c_1^{ref} should be a universal constant for all glasses as long as their viscosity exhibits a VFT-type temperature dependence and the high-temperature limit of their viscosity η_0 is 4×10^{-5} Pa·s. This is why, $c_1^{\text{ref}} = 17.44$ reported by Williams et al.⁴ is close to this universal value. On the other hand, $c_2^{\text{ref}} = T_g^* - T_0$ is determined by the relationship between T_g^* and T_0 . $c_2^{\text{ref}} = 51.6$ K observed by Williams et al.⁴ suggests that the ideal glass-transition temperature of the polymers and molecular glasses investigated therein is universally 51.6 K below their T_g .

Hence, it is seen that to examine the validity of the universal WLF formulation in MGs is to examine the values of $c_1^{\text{ref}} = D \cdot T_0 / \ln 10 (T_g^* - T_0)$ and $c_2^{\text{ref}} = T_g^* - T_0$ for MGs. For this purpose, the values of η_0 , D , T_g^* , and T_0 , for 89 MGs obtained in the past three decades by fitting the temperature-dependent viscosity and structural relaxation time with the VFT equation are carefully collected and analyzed. For specific values of these data of MGs, see Table S1. The data of MGs of which the relaxation time τ was measured are annotated. The η_0 value of these MGs is derived from the value of τ_0 via $\eta_0 = \tau_0 \cdot G_{\infty}$, where $G_{\infty} = 10^{10}$ Pa. Note that, as shown in the Supporting Information, various methods, such as beam bending,²⁵ parallel-plate rheometry,²⁶ levitation methods,²⁷ tensile tests,²⁸ methods based on crystal growth,²⁹ oscillating vessel methods,³⁰ etc., have been adopted to obtain the viscous dynamics of MGs. It is clear that the accuracy of the data would be highly dependent on the method adopted and affect the final results. In the current work, the fragility data are directly extracted from the literature and the potential error induced by the methods adopted in the literature are not distinguished.

3. RESULTS

Figure 1a,b shows the D vs T_g^* plot and η_0 vs T_g^* plot for various MGs, respectively. To get more detailed information on the source of the data, see Table S1 in the Supporting Information. In Figure 1a, the value of D of a wide variety of MGs spreads over a range of 2–50, indicating that both strong and fragile MGs are considered and that these data are representative for MGs. In Figure 1b, although considered a constant, a vastly varying η_0 from 10^1 to 10^{-6} Pa·s for MGs is shown. This is because of the different initial values chosen for η_0 in the data fitting procedure. An example is shown in Figure 2, the viscosity data covering a wide range of 10^5 – 10^{13} Pa·s, i.e., 8 orders of magnitude, for a $\text{Zr}_{46.75}\text{Ti}_{8.25}\text{Cu}_{7.5}\text{Ni}_{10}\text{Be}_{27.5}$

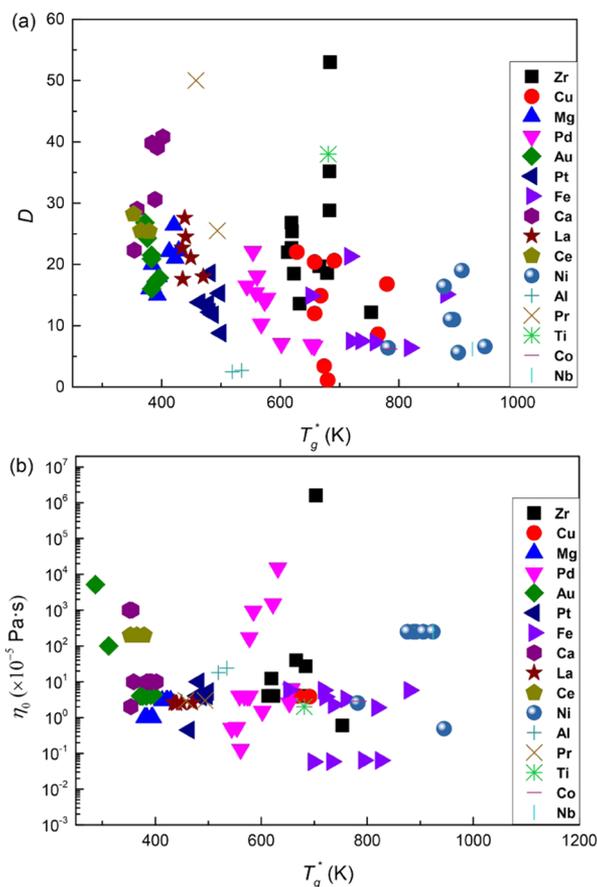


Figure 1. Compiled fragility data for various metallic glasses. (a) D vs T_g^* plot and (b) the η_0 vs T_g^* plot. D is the fragility parameter derived from the Vogel–Fulcher–Tammann (VFT) equation representing the deviation of the temperature dependence of viscosity from the Arrhenius relation; T_g^* is the temperature at which the viscosity equals 10^{12} Pa·s; η_0 is the high-temperature limit of viscosity. For more detailed information on the source of the data, see Table S1 in the Supporting Information.

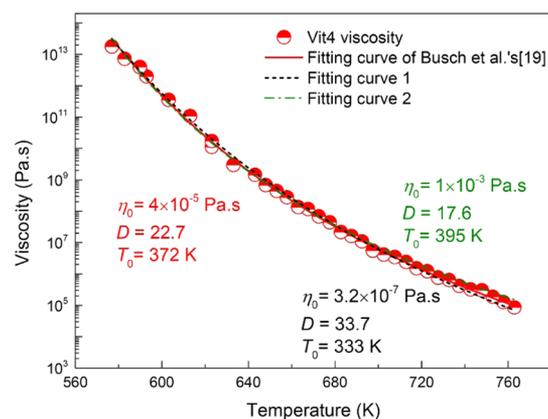


Figure 2. Example for the fits of the temperature-dependent viscosity data covering 8 orders of magnitude of a Vit4 metallic glass to the Vogel–Fulcher–Tammann (VFT) equation with different η_0 values.

(Vit4) MG was extracted from ref 19. For comparison, the fitting result provided by ref 19 was also given, i.e., $\eta_0 = 4 \times 10^{-5}$ Pa·s, $D = 22.7$, $T_0 = 372$ K. It is seen that the viscosity data could also be well fitted by other two groups of parameters of much different η_0 values, i.e., $\eta_0 = 1 \times 10^{-3}$

Pa·s, $D = 17.6$, $T_0 = 395$ K, and $\eta_0 = 3.2 \times 10^{-7}$ Pa·s, $D = 33.7$, $T_0 = 333$ K. This result suggests that with different initial values of η_0 (i.e., values near 10^{-3} , 10^{-5} , and 10^{-7} Pa·s), the fitting results could be quite different. Therefore, as explained in the **Methods** section, for valid values of D and T_0 , the reasonable values of $\eta_0 = N_A h / V$ in this work were chosen as of an order close to 10^{-5} Pa·s. Note that different values of η_0 have also been adopted in the literature.^{17,31}

Figure 3a shows the value of $\beta = c_1^g \ln 10$ for MGs of which the value of η_0 is of an order near 10^{-5} Pa·s. The data are close

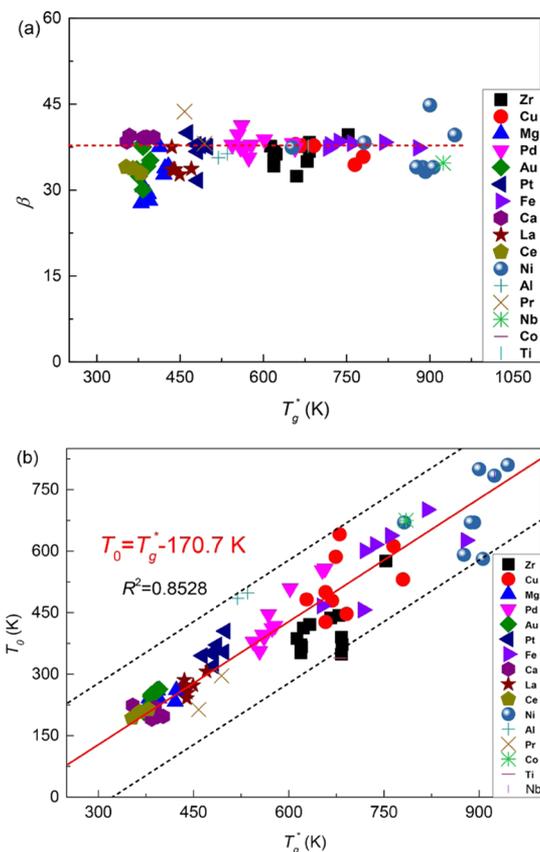


Figure 3. Universal scaling in metallic glasses. (a) Nearly constant β the short dashed line indicates the value of 37.75. (b) The nearly linear correlation between the glass-transition temperature T_g^* and the ideal glass-transition temperature T_0 for different metallic glasses (see the **Supporting Information** for more details). The solid line and the short dashed lines correspond to the linear equations with intercepts of 170.7 and 170.7 ± 150 K respectively.

to the constant 37.75 indicated by the dashed line suggesting the universal feature of c_1^g , whereas the dispersion of the data indicates the variation of η_0 and the experimental error in measuring viscosity. Figure 3b shows the correlation between T_g^* and T_0 for MGs, the values of which are directly obtained from the literature (see the **Supporting Information** for more details). Intriguingly, although with obvious fluctuations which overwhelm the experimental error between T_g^* and T_g , it is seen that T_g^* and T_0 exhibit a clear linear correlation, which could be approximated with $T_0 = T_g^* - 170.7$ K. This fact indicates that $c_2^g = T_g^* - T_0 \approx 170.7$ K and nearly maintains a constant for various kinds of MGs. Hence, $c_1^g = 16.4$ and $c_2^g \approx 170.7$ K suggest a universal scaling in the viscous dynamics of MGs as

$$\log \frac{\eta(T)}{\eta(T_g^*)} = \frac{-16.4(T - T_g^*)}{170.7 + (T - T_g^*)} \quad (14)$$

Since the fragility $m = \frac{c_1^g}{c_2^g} \cdot T_g^*$, for the roughly constant values of c_1^g and c_2^g of MGs, our results suggesting a crude linear correlation between m and T_g for MGs are consistent with previous works.³²

As to the relatively large error (± 150 K) shown in Figure 3b, it could be attributed to the following three aspects. First, the error could arise from the accuracy in the measurement of viscosity, relaxation time, and T_g of MGs. As stated previously, various methods, such as beam bending, parallel-plate rheometry, etc., have been adopted to obtain the viscous dynamics of MGs. The accuracy of the viscosity data would be highly dependent on the method adopted and lead to certain error in the value of c_2^g . Second, the error could also arise from the estimated value of η_0 and the data fitting procedure, which alter the result of c_2^g by affecting the fitting result of T_0 , as explained in the previous section. Finally yet most importantly, we argue that the relatively larger error mainly comes from the chemical short-range order in different MGs,³³ which fundamentally affects the viscous dynamics of MGs. However, detailed contributions of these three factors to the total error are an open question and require future investigation.

4. DISCUSSION

$c_2^g \approx 170.7$ K for MGs is more than three times higher than 51.6 K for polymers and molecular glasses. This difference could be rationalized from the aspect of the free volume theory,⁴ wherein the derivation of the WLF formulation is as follows.

$$\eta(T) = \eta_0 \cdot \exp\left(\frac{B_f \cdot v_0}{v_f}\right) \quad (15)$$

In eq 15, v_0 is the occupied volume, v_f is the free volume, and B_f is a material-dependent constant. The temperature dependence of free volume v_f could be approximated with: $v_f/v_0 \approx v_f/(v_0 + v_f) = (\alpha_l - \alpha_g)(T - T_g) + f_g$, where $f_g = v_f/(v_0 + v_f)|_{T=T_g}$, α_l and α_g are the thermal expansion coefficients of the equilibrium melt and the glass, respectively. Based on eq 15, by substitution and reorganization

$$\log \frac{\eta(T)}{\eta(T_g)} = \frac{-(1/\ln 10) \cdot B_f \cdot f_g^{-1} \cdot (T - T_g)}{f_g / (\alpha_l - \alpha_g) + (T - T_g)} \quad (16)$$

It is seen that $c_1^g = (1/\ln 10) \cdot B_f \cdot f_g^{-1}$ and $c_2^g = f_g / (\alpha_l - \alpha_g)$ and the physical meanings of the two coefficients are not explicit. Nevertheless, the values of c_1^g and c_2^g could be estimated. f_g is close to the volume change in MGs after crystallization and of an order of 3×10^{-3} .³⁴ $\alpha_l - \alpha_g$ is of an order of $2 \times 10^{-5} \text{ K}^{-1}$ for MGs.³⁵ Hence, a value of 150 K close to 170.7 K could be estimated for c_2^g . While for polymers,⁴ the values are 2.5×10^{-2} and $5 \times 10^{-4} \text{ K}^{-1}$, respectively, leading to a value of $c_2^g \approx 50$ K close to 51.6 K. It can be seen that the larger c_2^g for MGs is because of the smaller difference between the thermal expansion coefficients of metallic melt and MGs, and related to the volume change in MGs after crystallization. However, the volume change in MGs after crystallization is much smaller than that in polymers, and should lead to a smaller c_2^g . Thereby, we conclude that the larger c_2^g for MGs than that of polymers is

primarily attributed to the smaller difference between the thermal expansion coefficients of metallic melts and MGs. The difference in thermal expansion coefficients is very likely arising from the atomic configuration evolution of metallic liquids from equilibrium melt to glass, i.e., of a configurational origin, and implies the crucial role of configurational entropy in glass formation. As to c_1^g , the universal value 16.4 and a volume change of 3×10^{-3} requires that the constant B_f for MGs is about 0.1, in contrast to the B_f for polymers, which is of an order of unity.⁵ The much smaller B_f for MGs could be rationalized as follows. In free volume theory,⁵ the parameter B_f could be understood as a numerical correcting factor for the overlap of free volume. It is assumed to be between 1/2 and 1 for hard sphere systems, wherein larger B_f indicates less overlap of free volume and vice versa. As suggested by Cohen and Turnbull,⁵ for the metallic bonds between metal atoms, the atomic core of metal atoms plays the role of hard spheres and the critical free volume size is about 10% of the atomic volume of metal atoms, while this size in hard sphere systems is 80% of the volume of hard spheres, indicating that the free volume size in MGs is relatively smaller. Moreover, as found in MG models with a pairwise potential, large atomic size voids are unstable and break into smaller voids.³⁴ Thereby, the free volume in MGs is considered as distributed voids. From this perspective, it is likely that the correcting factor for the overlap of free volume in MGs is much smaller and indicates that the free volume in MGs is highly overlapped. Nevertheless, more specific explanations on the smaller value of B_f for MGs still require further investigation.

To further reveal the implications of this universal scaling in MGs and unravel how the coefficients $c_1^g = D \cdot T_0 / \ln 10 (T_g^* - T_0)$ and $c_2^g = T_g^* - T_0$ relate to glass formation, it is noted that the VFT equation could be derived from the Adam–Gibbs relation.⁶

$$\eta(T) = \eta_0 \cdot \exp\left(\frac{B}{T \cdot S_c}\right) \quad (17)$$

In eq 17, S_c is the configurational entropy and B is a material-dependent constant. Since S_c is dependent on the heat capacity difference ΔC_p between the equilibrium melt and the glass, assuming a hyperbolic form for ΔC_p : $\Delta C_p = \frac{C}{T}$ as having been observed in various MGs³⁶ with C being a material-dependent constant, $S_c(T) = \int_{T_0}^T \frac{\Delta C_p}{T} dT = C \cdot \frac{T - T_0}{T \cdot T_0}$. Equation 17 evolves into a VFT-type equation¹⁷

$$\eta(T) = \eta_0 \cdot \exp\left(\frac{C^{-1} \cdot B \cdot T_0}{T - T_0}\right) \quad (18)$$

In eq 18, $C^{-1} \cdot B = D$. As presented in eq 11, the WLF formulation could yet be derived from the VFT equation, and moreover, with T_g^* as the reference temperature, $c_1^g = D \cdot T_0 / \ln 10 (T_g^* - T_0)$ is a universal constant 16.4 for all glasses and $c_2^g = T_g^* - T_0$. According to the Adam–Gibbs relation,⁶ T_0 is the ideal glass-transition temperature at which configurational entropy vanishes. Therefore, the nearly constant c_2^g for MGs suggests that configurational entropy would vanish universally for all metallic glass-forming liquids after a further decrease in temperature of about 170.7 K below T_g^* . This phenomenon implies that configurational entropy probably decreases with temperature through a universal mechanism in the vitrification of MGs. However, to elucidate the detailed decreasing

mechanism requires advanced understanding on the origin of configurational entropy³⁷ and further investigations by molecular simulations.¹² Similar universality has also been suggested by molecular simulations where a quasi-universal entropy scaling in the diffusion of liquids was observed.³⁸

Notwithstanding being observed in polymers⁴ (Figure S1) and MGs, the nearly linear correlation between T_g^* and T_0 , which leads to a constant c_2^g , was not observed in oxide glasses^{32,39} (Figure S2). As argued by Tong et al.,^{11,12} configurational entropy resides in the packing capability of local particle arrangement and exhibits a many-body feature. It is thereby speculated that it is the strong orientational short-range order in the covalent network structure of oxide glasses⁴⁰ that intervenes the configurational entropy-dominated glass formation. This proposition is also in agreement with the above explanation that the relatively larger error of c_2^g mainly comes from the chemical short-range order in different MGs. In total, recalling the relatively simple molecular structures of metallic liquids,⁴¹ our work also indicates that MGs provide an ideal research subject for studies on the nature of glass formation.

5. CONCLUSIONS

In summary, a universal scaling in the temperature-dependent viscous dynamics of MGs is revealed in terms of the WLF

equation: $\log \frac{\eta(T)}{\eta(T_g^*)} = \frac{-c_1^g(T - T_g^*)}{c_2^g + (T - T_g^*)}$. In this formulation, $c_1^g = 16.4$

is proved to be nearly a universal constant for all glasses, and close to the value 17.44 observed by Williams et al. in polymers and molecular glasses; $c_2^g = 170.7$ K, which is larger compared to the value 51.6 K observed by Williams et al. in polymers and molecular glasses, is primarily attributed to the smaller difference between the thermal expansion coefficients of metallic melts and MGs. The universal WLF formulation, i.e., the nearly constant values of $c_2^g = 170.7$ K for MGs, indicates a universal vitrification mechanism mediated by configurational entropy in MGs.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.jpcc.1c00034>.

Complied data on the fragility of 89 metallic glasses (Table S1); T_g and T_0 in polymers and molecular glasses (Figure S1); and T_g and T_0 in oxide glasses (Figure S2) (PDF)

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Notes

The authors declare no competing financial interest.

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