



Mechanical failure and glass transition in metallic glasses

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ABSTRACT

The current majority view on the phenomenon of mechanical failure in metallic glasses appears to be that it is caused by the activity of some structural defects, such as free-volumes or shear transformation zones, and the concentration of such defects is small, only of the order of 1%. However, the recent results compel us to revise this view. Through molecular dynamics simulation it has been shown that mechanical failure is the stress-induced glass transition. According to our theory the concentration of the liquid-like sites (defects) is well over 20% at the glass transition. We suggest that the defect concentration in metallic glasses is actually very high, and percolation of such defects causes atomic avalanche and mechanical failure. In this article we discuss the glass transition, mechanical failure and viscosity from such a point of view.

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1. Introduction

Because deformation of crystalline materials occurs due to motion of dislocations, search for similar “defects” in metallic glasses has been attempted for a long time. Free volume [1] and shear transformation zone [2] are well-known examples of proposed defects. The concentrations of such defects are assumed to be low, as those of dislocations. For instance in the glassy state the free volume amounts only to the order of a percent in volume fraction.

However, these views are strongly challenged by recent results which suggest a very different global picture of the local structure of glasses. In the new picture defects are ubiquitous and occupy as much as a quarter of the volume. They cause local deformation which results in anelasticity, and plastic deformation occurs when the local anelastic deformations percolate through. In this paper we first discuss the nature of the defects in metallic glasses, and point out that it is dangerous to estimate its density from the volume change. We then review the result of a computer simulation which connects mechanical failure directly with glass transition [3] and the theory of the glass transition based upon the local topological fluctuations [4]. We finally discuss the role of defects in the anelastic behavior, and conclude with the proposal to define structural defects in metallic glasses in such a way to involve as much as a quarter of all atoms in the glass.

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2. Nature of structural defects in metallic glasses

In crystals lattice defects are unambiguously identified as deviations from the perfect lattice structure. In liquids and glasses, on the other hand, defects are hard to identify because there is no reference structure to define the defects against. Nevertheless the concept of defect is useful in describing the mechanical response. The variation in the local structure is the principal feature of the glassy state, so that the local mechanical response also varies from place to place. Thus it is natural to consider the portions of the system which respond strongly to the applied stress as defects. However, direct experimental observation of such defects is difficult.

Another phenomenon which is easier to comprehend in terms of defects is the structural relaxation. The hallmark of the liquid state is that its structure changes with temperature, although it is difficult to define exactly how it is changing. The structure obtained by rapid quenching from the liquid state depends on the temperature of the initial state, or the fictive temperature, T_f [5]. If the glass is annealed at a temperature, T_a , which is below the glass transition temperature, T_g , the structure will relax toward the equilibrium state at T_a . In other words T_f relaxes toward T_a . During the structural relaxation nearly all the properties of the glass change [6]. It is natural to explain the structural relaxation in terms of annealing out structural defects. Most often the free-volume model is invoked, because the volume is reduced by 0.5% or so during the structural relaxation.

However, it is dangerous to attribute the relaxation solely to the reduction in free-volume. During the relaxation heat is released. In

the free-volume theory the ratio of the released enthalpy, ΔH , to the volume change, ΔV , $\Delta H/\Delta V$, should be comparable to that of a vacancy in the solid, which is 2–3 eV per atom. But the actual value of $\Delta H/\Delta V$ measured during the structural relaxation is too large compared to what is expected by a factor of 5–10 [7]. A natural way to resolve this conundrum is to consider not only the defects where the local density is low (free-volume, or negative density n -type defect), but also the defects where the local density is high (anti-free-volume, or p -type defect) [8]. During the relaxation annihilation of free-volume reduces the total volume, but the annihilation of the anti-free-volume increases it. These changes cancel each other in the first order, so the total change in volume, ΔV , is the second order effect, much smaller than the actual local volume changes involved [7]. The change in the atomic pair-density function (PDF) due to the relaxation observed by X-ray diffraction [9] is explained well by assuming both positive and negative density fluctuations are eliminated by relaxation [10].

This duality of free-volume and anti-free-volume reflects the fact that the interatomic potentials in metallic glasses are more harmonic than those in other glasses such as polymeric glasses [11]. Thus the picture based upon the model of randomly packed hard-spheres [12,13] does not apply too well for metallic glasses, as was already pointed out by Cohen and Turnbull in their original paper published in 1959 [1]. They showed that the value of the critical free-volume, v^* , is a small fraction (about 10%) of the atomic volume for metallic liquids, whereas in the original free-volume theory it is supposed to be about 80% of the atomic volume. In addition, in the free-volume picture significant pressure dependence is expected for diffusivity, but for metallic liquids the measured pressure dependence is much smaller than expected for the free-volume model [14,15]. Also the isotope effect on diffusion is vanishingly small, again in contradiction to the free-volume model [16]. In metallic systems with harmonic potentials local density fluctuations occur both in positive and in negative directions. The intensity of such fluctuations cannot be measured by the density itself, but by the magnitude or amplitude of the local density fluctuations.

3. Equivalence of mechanical failure and glass transition

As is widely known metallic glasses mechanically fail by forming localized shear bands, and the fracture surface is characterized by the vein pattern which suggests local melting in the shear band [17]. Also there are ample reports of local heating within the shear band [18,19], which could reduce viscosity and lead to local melting. However, it is most likely that local heating is a consequence of rapid and severe deformation rather than the cause of deformation [19,20]. Instead of heating, local volume expansion, thus increased free-volume, was suggested as the cause of local deformation [20,21]. But it is highly questionable if local volume has time to expand in a very short time available for the nucleation of a shear band. Also the free-volume mechanism should result in a strong effect of pressure on deformation, but a careful experimental work by Varadarajan and Lewandowski [22] has shown that the effect of pressure on the fracture strength is remarkably small. They concluded that the fracture occurs when the shear stress reaches the critical value, and pressure has, if any, only minor effects. This result is consistent with the absence of the pressure effect on diffusion [14,15], and casts doubt on the free-volume mechanism of deformation.

Indeed the recent molecular dynamics (MD) simulation demonstrated that the applied shear stress has the equivalent effect as temperature in reducing the viscosity and inducing mechanical flow, even without local volume expansion [3]. In this work the steady-state flow under the stress, σ , was simulated for various

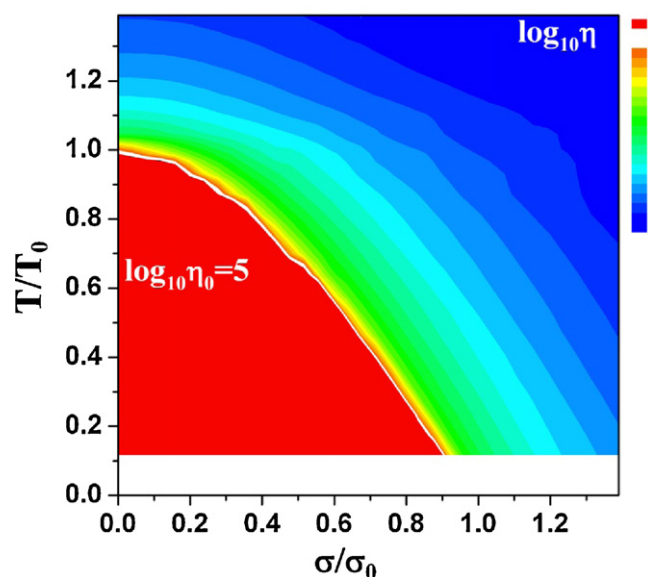


Fig. 1. The curves of equal viscosity as a function of normalized temperature and shear stress, computed by MD simulation on amorphous $Zr_{50}Cu_{40}Al_{10}$ [3]. The curves are self-similar, and can be collapsed into one universal curve.

temperatures, T , and strain rates, $\dot{\gamma}$, for a model system with a set of realistic potentials for $Zr_{50}Cu_{40}Al_{10}$ [23]. Both temperature and volume were kept constant during the simulation, so there was no effect of heating or dilation. The viscosity was calculated as $\eta = \sigma/\dot{\gamma}$. It was found that the shear stress alone is sufficient in reducing the viscosity and inducing the flow.

Moreover the constant viscosity curve plotted in the T - σ surface (Fig. 1 [3]) was found to be self-similar, and can be collapsed to a single curve by the surprisingly simple scaling relationship,

$$\frac{T}{T_0(\eta)} + \left(\frac{\sigma}{\sigma_0(\eta)}\right)^2 = 1 \quad (1)$$

where $T_0(\eta)$ is the T intersect ($\sigma=0$) of the curve for viscosity η , and $\sigma_0(\eta)$ is the σ intersect ($T=0$) of the same curve. The quadratic power of σ is consistent with the symmetry, because changing the sign of σ corresponds to shearing in the opposite direction, without changing the physics. The implications of this equation are discussed below. The result shown in Fig. 1 demonstrates the direct and intimate connection between the glass transition and mechanical failure, and shows that *mechanical failure is the glass transition induced by the applied stress*.

4. Atomistic mechanism of glass transition

Now that the nature of mechanical failure is established as the stress-induced glass transition, we need to understand the phenomenon of the glass transition itself. In our earlier publication we assumed that above T_g the atomic level volume strain, ε_v^T , follows the formula,

$$E_{el,LT} = \frac{VB \langle (\varepsilon_v^T)^2 \rangle}{2K_\alpha} = \frac{kT}{4}, \quad (2)$$

$$K_\alpha = \frac{3(1-\nu)}{2(1-2\nu)}$$

where B is the bulk modulus, V is the atomic volume, and ν is the Poisson's ratio [4]. The atomic level volume strain can be calculated from the atomic level pressure, p [24] as $\varepsilon_v = p/B$. Separately

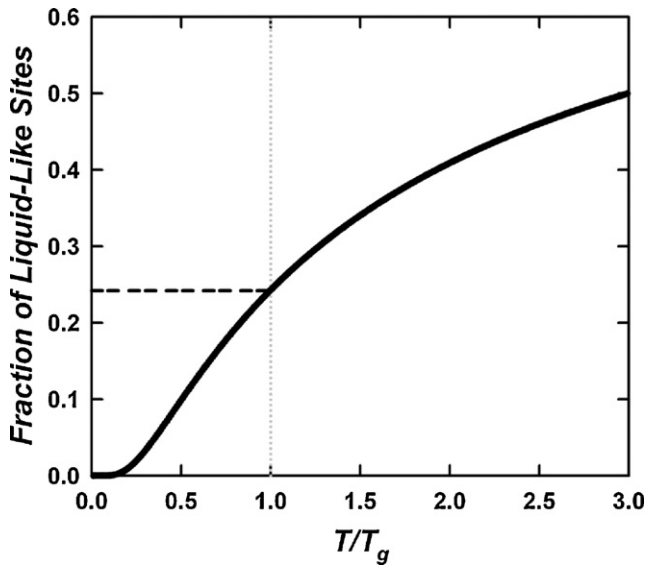


Fig. 2. The calculated fraction of the liquid-like sites, as a function of T/T_g . At T_g the fraction freezes at a constant value, as indicated by a dashed line.

we have shown that if the atomic level volume strain exceeds the critical volume strain,

$$|\varepsilon_v^{\text{crit}}| = 0.11 \quad (3)$$

the local topology, the arrangement of the nearest neighbor atoms, becomes unstable [25]. The atomic sites with the volume strain exceeding the value in Eq. (3) are defined as liquid-like defect sites. The density of the liquid-like sites, $p(\text{liq})$, depends on temperature, and is given by

$$p(\text{liq}) = CE(z) = \frac{2}{\sqrt{\pi}} \int_z^\infty e^{-x^2} dx, \quad (4)$$

$$z = \frac{\varepsilon_v^{\text{crit}}}{\sqrt{2} \langle (\varepsilon_v^T)^2 \rangle^{1/2}} = \frac{\varepsilon_v^{\text{crit}}}{\sqrt{K_\alpha kT/VB}}$$

where $CE(z)$ is the complementary error function [4]. Temperature dependence of $p(\text{liq})$ is shown in Fig. 2 as a function of T/T_g . The value of T_g used here was determined as below. Upon cooling $p(\text{liq})$ decreases with decreasing temperature, and when it reaches the critical concentration for percolation the glass transition should occur. Then $p(\text{liq})$ stays constant below T_g as shown by a dashed line in Fig. 2. Thus the glass transition temperature is given by,

$$kT_g = \frac{2BV}{K_\alpha} \left(\varepsilon_v^{T,\text{crit}} \right)^2. \quad (5)$$

The values of the glass transition temperature, experimentally determined for a large number of bulk metallic glasses, agree with Eq. (5) quite well with the value of the critical strain, $\varepsilon_v^{T,\text{crit}} = 0.095$. From Eqs. (5) and (6) we obtain,

$$z^2 = \frac{T_g}{2T} \left(\frac{\varepsilon_v^{\text{crit}}}{\varepsilon_v^{T,\text{crit}}} \right)^2 \quad (6)$$

This value gives $z = 0.825$ and $p(\text{liq}) = 0.243$ [4], which is approximately the percolation concentration for the dense random packed structure [26]. So the concentration of defects is not a few % as is widely assumed, but is of the order of 1/4. Indeed our recent X-ray diffraction study of a metallic glass showed that the volume fraction of defects is nearly 1/4, in excellent agreement with the estimates above [27]. In this work the structural change under the applied stress in the apparently elastic regime was examined by high-energy synchrotron X-ray diffraction using the anisotropic

pair-density function (PDF) analysis. It was found that a significant part of the mechanical response to the applied stress by a metallic glass is actually anelastic, even though the system behaves elastic macroscopically. The volume fraction of the anelastic sites was found to be 24%. In addition our theory is supported by the recent report that the jump in the specific heat is close to $(3/2)k_B$ per atom [28]. In our theory the configurational enthalpy above T_g is equal to $k_B T/4$ for each six component of the stress, adding up to $(3/2)k_B T$ in total, and freezes to a constant value below T_g . This produces the jump in the specific heat by $(3/2)k_B$.

5. Viscosity

Eq. (5) describes the equilibrium atomic configuration at T_g in terms of the average local strain, $\varepsilon_v^{T,\text{crit}}$, but does not describe the dynamics of structural excitations that determine viscosity and the kinetics of structural relaxation. In order to address this question let us examine how the atomic level strain is changed by local atomic rearrangement. The atomic level strain of an atom depends primarily on the topology of its nearest neighbor atomic cage [29]. Thus for the atomic level strain to change, the nearest neighbor cage has to undergo some collective displacements to alter its topology through a saddle point to a new stable configuration.

For instance the atomic level volume strain, ε_v^T , is directly related to the coordination number of that atom, N_C [29,30]. In order to relax the local volume strain of an atom i the local coordination number has to change. But N_C is an integer, so it can change only by unity at a time. Thus the strain which produces the change in N_C by 1/2 represents the saddle-point strain [25,31]. This logic was used in deducing the critical volume strain, Eq. (3). Then the local energy increase for the atom i is given by

$$\phi_{\text{saddle}} = \frac{BV}{2} \left(\varepsilon_v^{\text{crit}} \right)^2 \quad (7)$$

However, not only the i -th atom but also all the nearest neighbors also have to undergo the distortion collectively. Thus the total activation energy is multiplied by the cage effect,

$$E_a = A_{\text{cage}} \phi_{\text{saddle}} \quad (8)$$

The factor A_{cage} may be estimated for specific configurations [32], but in general it is difficult to calculate it directly. However, the activation energy at the glass transition temperature can be calculated using the expression for viscosity,

$$\eta = \eta_0 \exp(E_a/kT) \quad (9)$$

Because $\eta_0 = 10^{-4}$ poise [33] and $\eta = 10^{13}$ poise at T_g by definition, we obtain, at $T = T_g$,

$$E_a = CkT_g, \quad C = 39.1 \quad (10)$$

From Eq. (5),

$$\phi_{\text{saddle}} = \frac{K_\alpha kT_g}{4} \left(\frac{\varepsilon_v^{\text{crit}}}{\varepsilon_v^{T,\text{crit}}} \right)^2 \quad (11)$$

then,

$$A_{\text{cage}} = aC = 50.8, \quad a = \frac{4}{K_\alpha} \left(\frac{\varepsilon_v^{T,\text{crit}}}{\varepsilon_v^{\text{crit}}} \right)^2 = 1.30 \quad (12)$$

Above T_g A_{cage} is usually dependent upon $p(\text{liq})$, and thus T , because the presence of liquid-like atoms within the cage could make the cage weaker. This dependence is related to the fragility [33]. For a very strong liquid A_{cage} is independent of $p(\text{liq})$, thus of temperature. This can happen if the liquid-like sites are well separated from each other, and the cage does not include the liquid-like sites. In network glasses and polymers the weak links are van der

Waals bonds, whereas the strong covalently bonded network can separate the weak links from each other.

In random metallic glasses liquid-like sites can be next to each other. Then inclusion of the liquid-like sites in the cage decreases A_{cage} . Increase in temperature, thus in $p(\text{liq})$, will result in fast decrease in A_{cage} , and the system shows a fragile behavior. The fragility is related to the degree with which the liquid-like sites are incorporated in the cage. Strong chemical short-range order will prevent the inclusion of the liquid-like sites into the cage, and result in a strong behavior [31]. In general,

$$A_{\text{cage}}(T) = a \frac{E_a}{kT_g} = \frac{a}{x} \ln \frac{\eta}{\eta_0}, \quad x = \frac{T_g}{T} \quad (13)$$

With the fragility coefficient defined by

$$m = \frac{d \log \eta}{d(T_g/T)} \quad (14)$$

$$\frac{dA_{\text{cage}}}{dx} = -\frac{a}{x^2} \ln \frac{\eta}{\eta_0} + \frac{a'm}{x}, \quad a' = 2.3a \quad (15)$$

At $T = T_g$,

$$\left. \frac{dA_{\text{cage}}}{dx} \right|_{x=1} = a'(m-17), \quad \left. \frac{dA_{\text{cage}}}{dT} \right|_{T=T_g} = -\frac{a'(m-17)}{T_g}. \quad (16)$$

Now,

$$\frac{dp(\text{liq})}{dz} = -\sqrt{\frac{2}{\pi}} e^{-z^2/2} \quad (17)$$

$$\frac{dp(\text{liq})}{dx} = -\frac{z}{2x} \sqrt{\frac{2}{\pi}} e^{-z^2/2} \quad (18)$$

Thus at $T = T_g$,

$$\left. \frac{d \ln A_{\text{cage}}}{dp(\text{liq})} \right|_{T=T_g} = -C'(m-17), \quad C' = 0.111 \quad (19)$$

If we assume that A_{cage} changes linearly with the fraction of the solid-like sites, $p(\text{sol}) = 1 - p(\text{liq})$, for a totally random system $d \ln A_{\text{cage}} / d \ln p(\text{sol}) = 1$. This gives $m = 29$.

6. Mechanism of deformation

We now discuss the mechanism of deformation below T_g and the implication of the scaling relationship, Eq. (1). Below T_g A_{cage} is frozen to a constant value. If we apply an external shear stress, σ_{ext} , the distribution of the atomic level shear stress, $P(\sigma)$, becomes shifted to $P(\sigma + \sigma_{\text{ext}})$. Here we assume σ_{ext} is represented by only one of the five shear stress components, for instance σ_{xz} . Then the number of the atomic sites with the shear stress beyond the critical point, σ_{crit} , increases with σ_{ext} . As in Eq. (4) the density of the liquid-like site defined by the shear instability is given by

$$p_{\text{liq},s}(z, \delta) = \frac{1}{\sqrt{2\pi}} \left[\int_{z+\delta}^{\infty} e^{-x^2} dx + \int_{\infty}^{-z+\delta} e^{-x^2} dx \right] \quad (20)$$

$$z = \sigma_{\text{crit}} \sqrt{\frac{V}{K_\gamma G k T_g}}, \quad \delta = \sigma_{\text{ext}} \sqrt{\frac{V}{K_\gamma G k T_g}}, \quad K_\gamma = \frac{15(1-\nu)}{7-5\nu}$$

Expanding by δ we obtain,

$$p_{\text{liq},s}(z, \delta) = p_{\text{liq},s}(z, 0) + 2\sqrt{\frac{2}{\pi}} z e^{-z^2} \delta^2 + \dots \quad (21)$$

From Eq. (19),

$$\frac{dA_{\text{cage}}(\delta)}{A_{\text{cage}}(0)} = -C'(m-17) dp_{\text{liq},s}(z, \delta) = -C''(m-17) z e^{-z^2} \delta^2 + \dots \quad (22)$$

$$C'' = 2\sqrt{\frac{2}{\pi}} C' = 0.178$$

With Eq. (8),

$$E_a = A_{\text{cage}}(0) \phi_{\text{saddle}} \left[1 - C''(m-17) z e^{-z^2} \delta^2 \right]$$

$$= A_{\text{cage}}(0) \phi_{\text{saddle}} \left[1 - \left(\frac{\sigma_{\text{ext}}}{\sigma_0} \right)^2 \right], \quad \sigma_0 = \frac{\sigma_{\text{crit}}/z}{\sqrt{C''(m-17) z e^{-z^2}}} \quad (23)$$

The scaling relation, Eq. (1), can indeed be rewritten with the aid of Eq. (9) in the same form as Eq. (23) [4]. The effect of the external stress is to increase the density of the liquid-like sites through Eq. (21) and to reduce the cage factor, thus the activation energy through Eq. (23).

7. Conclusions

During the structural relaxation the volume decreases by about 0.5%, and at the same time various properties undergo significant changes. This observation is easy to understand in the framework of the free-volume theory, which has been widely used in the field of metallic glasses in spite of the warning in the original paper [1]. Thus the majority of researchers subscribe to the idea that the density of structural defects in metallic glasses is of the order of 1%. On the other hand in the high-temperature liquid state all atoms are unstable. Therefore one might conclude that a significant fraction of atoms belong to the defects. Thus in the free-volume theory during supercooling and cooling through the glass transition the majority of the defects are eliminated. In the Adam-Gibbs theory [34] as well this drastic decrease in the defect density was directly related to the precipitous decrease in the configurational entropy during cooling. In both theories in the almost immobile glassy state the defect density is quite low. On the other hand, in the percolation theory of Cohen and Grest [35] and in our theory [4] the defect density needs to come down only to the percolation concentration, which is not a few %, but is of the order of 20%. Below T_g defects are not eliminated, but they are localized and contribute only to anelasticity. Recent data [27] are in support of the high density of defects. In addition our recent simulation [3] directly connects mechanical failure and the glass transition. Thus mechanical failure as well is caused by percolation of defects with substantial densities, and not by a small number of topological defects which play the equivalent role of dislocations in crystals. The accumulation of new evidences are making a compelling case for the view that concentrations of defects in glasses are actually quite high.

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