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Ductile-to-brittle transition in spallation of metallic glasses

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In this paper, the spallation behavior of a binary metallic glass Cu50Zr50 is investigated with molecular dynamics simulations. With increasing the impact velocity, micro-voids induced by tensile pulses become smaller and more concentrated. The phenomenon suggests a ductile-to-brittle transition during the spallation process. Further investigation indicates that the transition is controlled by the interaction between void nucleation and growth, which can be regarded as a competition between tension transformation zones (TTZs) and shear transformation zones (STZs) at atomic scale. As impact velocities become higher, the stress amplitude and temperature rise in the spall region increase and micro-structures of the material become more unstable. Therefore, TTZs are prone to activation in metallic glasses, leading to a brittle behavior during the spallation process. © 2014 AIP Publishing LLC [http://dx.doi.org/10.1063/1.4897552]

I. INTRODUCTION

Due to the unique disordered microstructures, metallic glasses (MGs) have many excellent properties and receive much attention in recent years.1–8 It is well known that metallic glasses usually exhibit a brittle behavior like a glass at macroscopic scale, but show different capability of plastic deformation at microscopic scale.9–13 Thus, two distinct morphologies are usually observed on the fracture surfaces of MGs. For brittle fracture, the fracture surfaces are flat with nano-scale periodic corrugations or dimple structures;13–15 but for ductile fracture (not globally), much coarser patterns are found, such as river-like and cellular patterns as well as honeycomb structures.16,17

The fracture behavior of MGs is sensitive to their composition, and Mg-based and Fe-based MGs are usually much more brittle than Zr-based MGs.13,18,19 The fabrication process is also important. The longer the annealing time is, the more brittle MGs are.3 Besides, different loading conditions may lead to various fracture behaviors. During plate-impact experiments, Gupta and coworkers20,21 found that spallation of a Zr-based MG exhibits a ductile-to-brittle transition. With increasing the impact velocity, the pull-back velocity slope increases monotonically, which indicates that the loading-unloading response of the MG at macroscopic scale is more brittle. Further examination show that the spalled surfaces at microscopic scale agree with the macroscopic phenomenon.21,22 Smoother morphologies are observed at a higher impact velocity, while much coarser patterns are observed at a lower impact velocity.

To answer the question of what controls the ductile-to-brittle behavior in MGs, extensive works have been made over the past decades. On one hand, some researchers tried to find the macroscopic mechanical parameters that dominate the ductile-to-brittle transition process. In 1975, Chen et al.23 found that Poisson’s ratio is closely correlated with plasticity of MGs. Schroers and Johnson24 further proved that the larger the Poisson’s ratio, the better is the plasticity of MGs. Equivalent with Poisson’s ratio, another parameter μ/κ revealed by Lewandowski et al.25 is a key parameter controlling the ductile-to-brittle transition of MGs, where μ is the shear modulus representing the resistance to plastic deformation, and κ is the bulk modulus or the resistance to dilation. A lower μ/κ or larger Poisson ratio implies more ductile behavior. It is noted that MGs usually exhibit a significant tension-compression plasticity asymmetry and shear-induced dilation. Considering these intrinsic characters, Chen et al.10,11 recently took the intrinsic strength of the material into consideration, and proposed a shear-to-normal strength ratio and a strength-differential factor to characterize the ductile-to-brittle behavior in MGs. A smaller implies enhanced plasticity, while a larger indicates brittle fracture under tensile loading. On the other hand, researchers intended to find the answer at atomic scale. Based on a overview of fracture patterns, Jiang et al.14 argued that the ductile-to-brittle transition of MGs is controlled by competition between shear transformation zones (STZs)26–28 and tension transformation zones (TTZs)9,14,21,29 at microscopic scale. In contrast to STZs that are corresponding to shape distortions of atomic clusters under shear stresses, TTZs are regarded as the fundamental carriers of bulk dilations under negative pressures.7,14 When TTZs dominate, more brittle fracture behavior is expected. This view is supported by recent impact toughness tests21 and spallation experiments.9 In these tests, typical brittle fracture patterns are observed such as nm-sized vein patterns21 and nanosized corrugations,7 and TTZs are thought to be the reason for the phenomenon. More recently, Murali et al.20,31 studied the fracture behavior of two typical MGs (FeP and CuZr) via...
atomistic simulations. It is revealed that even a brittle fracture is dominated by nucleation and growth of voids in MGs, and a higher degree of spatial fluctuation induces more brittle behavior during the fracture process. Despite extensive investigations, the atomistic scale mechanism that governs the ductile-to-brittle transition in MGs is still unclear.

To reveal the ductile-to-brittle transition mechanism during a spallation process, we present molecular-dynamic (MD) simulations of a binary MG Zr\textsubscript{50}Cu\textsubscript{50} in this paper. By using a flyer-target configuration, the spallation behavior is studied at different impact velocities from 600 m/s to 1800 m/s, with emphasis on the damage evolution process. It is found that as the impact stress increases, a ductile-to-brittle transition occurs, which agrees well with the available experimental results. Further investigation reveals that the interaction between void nucleation and growth, which can be interpreted as the competition between TTZs and STZs at atomic scale, controls the ductile-to-brittle transition during the spallation process.

II. MD SIMULATIONS OF SPALLATION

During the MD simulations, a simple binary MG Zr\textsubscript{50}Cu\textsubscript{50} is selected as the model material. To model the atomic interactions in the Zr-Cu system, we adopt the Finnis-Sinclair type interatomic potential with parameters given by Mendelev et al.\textsuperscript{32} Calculations are carried out with the open source code LAMMPS.\textsuperscript{33} Glass samples are prepared via a melting-and-quenching process. The initial system is a fcc lattice with the sites randomly occupied by Zr and Cu atoms in accordance with the nominal composition. It consists of ~440,000 atoms arranged in a cubic shape, and three-dimensional periodic boundary conditions with ambient pressure are applied. To obtain the Zr\textsubscript{50}Cu\textsubscript{50} glass, simulations are performed in the constant number of particles, pressure, and temperature (NPT) ensembles with a time step of 1 fs. Temperature gradually increases from 1 K to 2500 K, equilibrates for 100 ps, and a glass sample is prepared with dimensions of ~20 \times 20 \times 20 nm\textsuperscript{3}.

In simulations of spallation, we construct the traditional flyer-target configurations.\textsuperscript{34,35} The flyer plate consists of ~200,000 atoms with dimensions of ~100 \times 20 \times 20 nm\textsuperscript{3}, and the target has the same cross-section area (20 \times 20 nm\textsuperscript{2}) but its thickness is twice as that of flyer. To obtain such a large system, the 400,000-atom glass (~20 \times 20 \times 20 nm\textsuperscript{3}) is replicated along the X direction, and equilibrates for another 100 ps to remove possible artifacts from the replication process.\textsuperscript{34} In fact, we have also explored the flyer-target system with a cross-section area of ~10 \times 10 nm\textsuperscript{2} to examine the size effect on spallation and the results are similar. In our simulations, the loading direction is along the X axis, so the nonimpact sides of flyer and target normal to the X axis are free surfaces. But along the Y and Z axes, the periodic boundary conditions are maintained to mimic one-dimensional (1D) strain shock loading. Here, we denote the desired impact velocity as V. The flyer plate and target are assigned initial velocities of 2V/3 and −V/3 before impacting, so that the flyer-target system has a center-of-mass velocity of 0. Shock simulations adopt the constant number of particles, volume, and energy (NVE) ensembles. The time step for integrating the equations of motion is 1 fs, and the run duration is 120 ps.

To obtain the physical properties of plates, the 1D binning analysis is used. The simulation cell is divided into fine bins along the X axis by neglecting the heterogeneities in the transverse directions, and we obtain the average physical properties such as density (ρ), stresses (σ\textsubscript{r}), particle velocity (u\textsubscript{p}), and temperature (T) profiles. To characterize the atomic configuration, we use the Voronoi tessellation analysis.\textsuperscript{36} And the plastic deformation is identified by the nonaffine displacement $D^2_{\text{min}}$ proposed by Falk and Langer.\textsuperscript{27}

III. RESULTS

During the shock simulations, the thickness of flyer plates and targets are not changed. To achieve shock loading with different amplitudes, we choose impact velocities $V$ of 600, 900, 1200, 1500, and 1800 m/s, respectively. Figure 1 illustrates the free surface velocity histories on the target side, similar to that measured by a velocity interferometer system for any reflector (VISAR) in plate-impact experiments.\textsuperscript{20,37,38} As shown in Fig. 1, typical “pull-back” waves, which are signatures for spallation, are observed in all cases, except for the case of $V = 600$ m/s. It indicates that spallation occurs in the cases of $V = 900$, 1200, 1500, and 1800 m/s. Besides, as the impact velocity increases, the pull-back velocity slope also increases. It agrees well with the experimental results,\textsuperscript{20} which indicates a ductile-to-brittle transition behavior. To compare the spallation behaviors under different loading amplitudes, the cases of $V = 900$ and 1500 m/s are further characterized.

Figure 2 shows the density evolution in a conventional $x$ - $t$ diagram at impact velocities of 900 and 1500 m/s. With color coding based on the local atomic number density, the wave propagation and interaction process is illustrated, which is related to the shock, release, tension, and spallation. As shown in Fig. 2, the red color represents regions with a higher density, while the blue color represents regions with a lower density. A deeper blue color implies a larger amount

FIG. 1. Free surface velocity histories on the target side at different impact velocities.
of spallation damage. According to the process of wave propagation, the tensile stress duration needed for spallation at the lower impact velocity is much longer than that at the higher velocity. It is obvious that the distribution of spallation damage is different between these two impact velocities. In the case of $V = 900$ m/s, damage is scattered over the spall plane. But in the case of $V = 1500$ m/s, it is more concentrated.

The corresponding stress profiles ($\sigma_x$) at different times before and after spallation at $V = 900$ and 1500 m/s are shown in Fig. 3. It is seen that the tensile region is formed due to the interaction of two release waves reflected from the free surfaces of flyer and target. As micro-damage nucleates and grows, recompression waves are generated in the spalled region and propagate toward the free surface. The recompression wave is registered in the free surface velocity profile as a “pull-back” wave. Compared with the case of $V = 1500$ m/s where there is only one recompression wave, two recompression waves are observed near the spall plane at $V = 900$ m/s, which imply that a multi-spall occurs. The result is in accordance with the scattered distribution of spallation damage at a lower impact velocity, as shown in Fig. 2.

Next, we examine the damage evolution process in the spalled region (where the recompression wave is generated) at different impact velocities. Figures 4(a)–4(c) show the spallation damage at the impact velocity of 900 m/s, and Figs. 4(d)–4(f) show the damage at $V = 1500$ m/s. As the impact velocity varies, the rate of damage evolution is different. Thus, in the case of 900 m/s, a time spacing of 5 ps is used to track the spall process, while 3 ps is adopted at $V = 1500$ m/s. As shown in Fig. 4, spallation of Cu$_{50}$Zr$_{50}$ glass undergoes the process of nucleation, growth, and coalescence of micro-voids. At a lower impact velocity, only a few large voids (actually only one in the slice) dominate the damage evolution process. In contrast, a large number of voids can be observed at a higher impact velocity. The voids are small and begin to coalesce. The damage characteristics imply a smoother morphology on the fracture surfaces at a higher impact velocity.

IV. DUCTILE-TO-BRITTLE TRANSITION MECHANISM

According to the results of plate-impact experiments, there are two typical characteristics at different impact velocities, which suggests a ductile-to-brittle transition during spallation of MGs: (1) at macroscopic scale, the pull-back velocity slope increases with increasing the impact velocity; and (2) at microscopic scale, it is frequent to observe a smoother morphology on the fracture surfaces of the spalled samples at a higher impact velocity, while much coarser patterns are observed at a lower impact velocity. Our results generally agree with the experimental results as shown in Figs. 1 and 3.

In the MD simulations, the most obvious difference between the fracture phenomena at different impact velocities is the change of generated void numbers. With increasing the impact velocity, there are much more voids observed to nucleate and grow on the spall plane. The larger the void number is, the smaller the void sizes are before coalescence. Then the fracture surface is smoother, which is a typical characteristic in brittle fracture. This interesting phenomenon has also been observed in other works. For example, during the MD simulations of the fracture behavior of two typical MGs (FeP and CuZr), more smaller voids are observed in brittle FeP MG, while one bigger void is found in ductile CuZr MG. As the fracture behavior (brittle or ductile) is determined by the plastic deformation at microscopic scale, this phenomenon implies that the plastic deformation is impeded when more voids are generated. Now the question...
is, why the plastic deformation is impeded in the case with more and smaller voids.

A. Competition of TTZs and STZs

In order to reveal the factors that influence the plastic deformation during spallation, we explore the process of void nucleation and growth. Figure 5 shows the nonaffine displacement during the void nucleation and growth at $V = 900$ m/s. Here, the critical size for void nucleation is determined to be $\sim 1$ nm in diameter. Thus, according to the void size, Figs. 5(a)–5(c) illustrate the nucleation process, and Figs. 5(d)–5(e) exhibit the growth process. As shown in Figs. 5(a)–5(c), during the void nucleation process, atoms with a larger nonaffine displacement are randomly distributed in the material. With increasing the time interval (the reference configuration is the same at $t = 81$ ps in Fig. 5), the number of atoms with a larger nonaffine displacement increases. There is no apparent difference observed between the void nucleation location and other region. It implies that the nonaffine displacement is induced by temperature (or structural relaxation) instead of stresses. However, during the void growth process, nonaffine displacement of atoms in the region around the void is much larger than that away from the void. It indicates that plastic deformation of the material is mainly induced by void growth, there is nearly no contribution from void nucleation.

FIG. 4. Damage evolution process at different impact velocities: (a)–(c) $V = 900$ m/s; and (d)–(f) $V = 1500$ m/s. The colors indicate the normalized local atomic number density.

FIG. 5. Snapshots of void nucleation and growth at $V = 900$ m/s: (a)–(c) Nucleation of voids; and (d)–(f) Growth of voids. The colors represent the value of $D^2_{\text{min}}$ calculated from the same reference configuration at $t = 81$ ps.
Further investigation reveals that the nucleation and growth of voids is closely related to the fundamental unit-processes of collective atomic motion in MGs. Figure 6 shows some close-up views of the atomic cluster motion around the void. As shown in Fig. 6(a), during the nucleation process, transformation of the atomic structure at the centre of the void is similar to the picture of a TTZ. But during the growth process as shown in Fig. 6(b), the motion of the atomic cluster at the edge of the void is close to the picture of a STZ. We know that TTZs are corresponding to bulk dilations of atomic clusters, but STZs arouse shape distortions (the accompanied dilations are very small). As STZs are mainly activated during the void growth process, plastic deformation induced by damage evolution in the material is attributed to the void growth process.

Now the question is, as the impact velocities increase, why does plastic deformation decrease? Because plastic deformation is closely related to the void growth process, void growth at different impact velocities is examined. We compare the diameter history of the biggest voids at $V = 900$ and $1500$ m/s, as shown in Fig. 7. At a lower impact velocity, we see that the void grows continuously with a gradually increasing growth rate. But at a higher impact velocity, the void grows fast at the initial stage, but the growth rate decrease a lot after a short time of ~4 ps. The difference can be explained by the damage evolution process as shown in Fig. 4. As there are more voids at the higher impact velocity, a growing void quickly interacts with the surrounding voids, leading to a coalescence process. This impedes the further growth of voids. However, as there is only one void at the lower impact velocity, it can grow continuously without confinements of other voids. Based on the above results, we think that the plastic deformation during spallation of MGs is controlled by competition of two rate processes at microscopic scale. On one hand, the void growth process promotes plastic deformation in the material. According to the conventional void growth mechanism, the plastic zone around the void is proportional to the void volume. Bigger voids induce a larger region of the material to undergo plastic deformation. Thus, the larger the voids grow, the more extensive plastic deformation the material undergoes. On the other hand, the void nucleation process impedes plastic deformation in the material. As void growth is bounded by the spacing between two nucleation sites, a higher nucleation rate which decreases the spacing between voids impedes the growth process. Therefore, plastic deformation in the material is slight.

In fact, the interaction between nucleation and growth can be interpreted as a competition between the fundamental unit-processes of collective atomic motion in MGs. Since void nucleation is related to the activation of TTZs, and growth is induced by STZs around the voids, the damage evolution process is intrinsically a competition between...
TTZs and STZs. To characterize the competition process, here we propose a non-dimensional number composed of two time scales:

\[ I_a = \frac{T_{STZ}}{T_{TTZ}} \]  

(1)

where \( T_{STZ} \) and \( T_{TTZ} \) are the characteristic time scales for activation of STZs and TTZs, respectively. As to a larger \( I_a \), TTZs are more dominant than STZs. To determine the two time scales, we estimated the activation rates of TTZs and STZs.

According to the STZ models, \(^{27,43}\) the activation rate of a single potential STZ is written as

\[ v_{STZ} = \frac{1}{T_{STZ}} = v_1 \exp \left( -\frac{\Delta F_1 - \tau \cdot \gamma_0 \cdot \Omega_0}{k \theta} \right), \]  

(2)

where \( v_{STZ} \) is the STZ activation rate, \( v_1 \) is an attempt frequency of order of the Debye frequency, \( \tau \) is the local shear stress, \( \gamma_0 \) is the characteristic shear strain with the order of \( \sim 0.1 \), \( \Omega_0 \) is the STZ volume, \( k \) is the Boltzmann constant, \( \theta \) is the temperature, and \( \Delta F_1 \) is the activation barrier.

For TTZs, they are similar in size to STZs, and are activated by high hydrostatic tensile pressure. In the same way, we can estimate the activation rate of a single TTZ as

\[ v_{TTZ} = \frac{1}{T_{TTZ}} = v_2 \exp \left( -\frac{\Delta F_2 - \rho \cdot e_v \cdot \Omega_0}{k \theta} \right), \]  

(3)

where \( v_{TTZ} \) is the TTZ activation rate, \( v_2 \) is an attempt frequency, \( \rho \) is the hydrostatic tensile pressure, \( e_v \) is the characteristic volumetric strain, and \( \Delta F_2 \) is the activation barrier, which is mainly related to the dissipated energy forming new surfaces.\(^1\) Thus,

\[ I_a = \frac{v_1}{v_2} \exp \left( \frac{\Delta F_1 - \tau \cdot \gamma_0 \cdot \Omega_0}{\Delta F_2 - \rho \cdot e_v \cdot \Omega_0} \right), \]  

(4)

where \( v_1, \Delta F_1, \gamma_0, \Omega_0 \) are material parameters according to STZ models. If \( v_2, \Delta F_2, \) and \( e_v \) are also regarded as material parameters, \( I_a \) is determined by local stress states.

Further analysis indicates that the local stress states change before and after voids are nucleated. When there is no void in the material during spallation, it is the 1D strain condition and the ratio of shear stress \( \tau \) to tensile pressure \( p \) is

\[ \frac{\tau}{p} = \frac{\mu}{\kappa}, \]  

(5)

where \( \mu \) is the shear modulus and \( \kappa \) is the bulk modulus. For Cu-50Zr, \( \mu = 22 \) GPa and \( \kappa = 123 \) GPa, therefore, \( \tau/p \approx 0.18 \). Since the shear stress is much smaller than the tensile pressure, TTZs may play a dominant role according to Eq. (4). However, after voids are nucleated, the local stress states are completely changed. Although the tensile pressure \( p \) is nearly the same, the ratio of \( \tau \) to \( p \) around the void increases to 0.75 (as a rough estimate, the asymmetry of the loading and initial void shape is not taken into account). Thus, \( I_a \) will decrease, and STZs may play a dominant role. Note that only around the void’s surrounding where stress concentration takes place, \( I_a \) is smaller. For the region that is not influenced by the void, \( I_a \) is still relatively large and TTZs is the dominant collective atomic motion.

**B. Mechanism resulting in dominance of TTZs**

If MGs undergo brittle spallation, it is obvious that TTZs must dominate the fracture process. According to Eq. (3), factors such as stresses, temperature, and the activation barrier can influence the activation of TTZs in the material. In order to find the reason that results in dominance of TTZs, we further consider the evolution of the above factors at different impact velocities.

Figure 8 shows a comparison of the stress profiles at the beginning of the damage evolution process. With increasing impact velocity, the stress amplitude near the spall plane is slightly higher. According to Eq. (3), a higher tensile stress can increase the work done by the system, and decreases the energy barrier of TTZs, thus a higher activation rate is expected. Besides, it should be noted that micro-inertia might have influence on the competition between STZs and TTZs. As the impact velocity increases, the loading rate is higher and micro inertial effects on void growth become more important. Activation of STZs around the voids may be impeded by micro inertial effects, leading to a decrease of void growth rate.

The history of material temperature near the spall plane is illustrated in Fig. 9(a). The temperature keeps constant at first, then increases sharply as the flyer impacts the target, and finally decreases a little when the region of tension is created. Compared with the case with \( V = 900 \) m/s, the material temperature is apparently higher at \( V = 1500 \) m/s. As higher temperature implies that atoms have a higher chance of getting enough energy from thermal fluctuation to overcome the free energy barrier, it contributes to a higher activation rate.

For the activation barrier, it is determined by local atomic structures at the potential TTZ sites. Here, the degree of local fivefold symmetry (LFFS) is used as a key factor to characterize the local atomic structures.\(^{44}\) In the Voronoi tessellation analysis, each atom is indexed with the Voronoi indices \( \langle n_3, n_4, n_5, n_6, \ldots \rangle \), where \( n_3, n_4, n_5, \) and \( n_6 \) represent...
the number of triangles, tetragons, pentagons, and hexagons on the Voronoi polyhedron, respectively, and the degree of LFFS is defined as the fraction of the number of pentagons ($LFFS = \frac{n_5}{\sum n_i}$). The average degree of LFFS in the region near the spall plane is shown in Fig. 9(b). It is clear that the average degree of LFFS increases under compression and decreases under tension at both impact velocities. But at the time just before voids begin to nucleate, the average degree of LFFS can decrease to lower amplitude at the higher impact velocity. Since a lower LFFS indicates that the structural configuration of atoms is packed more loosely and has higher potential energy, the local structure is more unstable and it is easier for transformation of local atomic clusters.

Further examination indicates that TTZs are prone to activation from the region with a lower average LFFS. As shown in Fig. 10, at the impact velocity of 900 m/s, the average degree of LFFS of the atomic cluster that void originates from is 0.422, while that of the entire slice near the spall plane is 0.472. As the impact velocity increases to 1500 m/s, the average degree of LFFS of the atomic cluster that the biggest void originates from (0.431) is also smaller than that of the entire slice (0.464). It indicates that TTZs are indeed easier to be activated in the region with a lower degree of LFFS. As a previous work has shown that STZs prefer to be initiated in regions with a lower degree of LFFS too, it is obvious that a lower degree of LFFS means a lower activation barrier for transformation of atomic clusters. As the average degree of LFFS in the spall region is smaller at a higher impact velocity, there are more potential sites for activation of TTZs. Thus, the activation rate is higher.

V. CONCLUSION

We have studied the ductile-to-brittle transition phenomenon during spallation of a binary MG Zr$_{50}$Cu$_{50}$ with MD simulations. Our results show that as the impact velocity increases, the distribution of spallation damage becomes more concentrated and the fracture patterns are smoother, which agrees well with experimental observations in recent works. The ductile-to-brittle transition in spallation is related to extra fracture energy dissipation at a lower impact velocity and impedance of plastic deformation at a higher impact velocity. Plastic deformation during the damage evolution process is controlled by the interaction of two microscopic rate processes (i.e., void nucleation and growth), which can be interpreted as the competition of STZs and TTZs at atomic scale. As the impact velocity increases, TTZs dominates the fracture process and spallation exhibits a brittle behavior. Further investigation shows that with increasing the impact velocity, the tensile stress amplitude and material temperature is higher in the spall region, and the atomic structure is more unstable. All these reasons induce a larger void nucleation rate or the dominance of TTZs.
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33LAMMPS: MD simulation code available from Sandi National Laboratories, USA.