



Effect of temperature on shear bands and bending plasticity of metallic glasses



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ABSTRACT

We report the effect of testing temperature on the bending plasticity of $\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$ and $\text{Zr}_{35}\text{Ti}_{30}\text{Cu}_{8.25}\text{Be}_{26.75}$ metallic glasses. Bend tests ranging from liquid nitrogen to the glass transition temperature were conducted using a customized setup. The bending strain, the shear band density, and the critical shear offset at fracture decreased with increasing temperature in both the metallic glass formers. The constant strain experiments (without fracture) revealed that low temperature generates more shear bands of smaller lengths to accommodate the same applied strain. The results can be rationalized in terms of temperature dependence predicted by the nucleation and propagation models.

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1. Introduction

Room temperature plasticity of metallic glasses is controlled by localized shear bands which are 10–20 nm thick [1–4]. Number of shear bands is considered as a good indicator of intrinsic plasticity and fracture toughness of a metallic glass [4–8]. Consequently, majority of recent research on metallic glasses has been directed towards understanding and controlling the shear bands. Effects of composition, structure, and processing on shear banding in metallic glasses have been extensively studied [4,9–11]. Alloys with a lower shear to bulk modulus ratio (or higher Poisson ratio) form multiple shear bands, and as a result, exhibit higher plasticity [12,13]. Structural features such as, free volume, icosahedral clusters, density fluctuations, and chemical heterogeneities have been correlated with enhanced shear band nucleation [5,14–16]. Higher cooling rate, shot-peening, and thermal rejuvenation also enhance propensity for shear band proliferation in metallic glasses [17–21]. In addition, the testing methodology, temperature, strain-rate, and sample size can strongly influence the shear banding process and overall plasticity of metallic glasses [22–29].

As earlier shown by Spaepen and confirmed by others, the effects of temperature and strain-rate on the flow behavior of metallic glasses can be summarized in a deformation map [2,30–32]. At low temperature or high strain-rate, the metallic

glasses deform through localized shear bands. At high temperature (close to glass transition) or extremely low strain-rate, localization is averted and metallic glasses deform homogeneously. These two flow regimes (localized and homogeneous) have been explained on the basis of competing time scales for structural relaxation and creation of disorder during deformation. However, the effect of temperature on shear band mediated plasticity in localized regime remains controversial. A significant enhancement in plasticity at cryogenic temperatures has been reported in several metallic glass formers [33–36]. Stress-strain curves become non-serrated at cryogenic temperatures while the plastic flow is still accommodated by multiple shear bands [35–37]. Some metallic glasses even become brittle again at very low temperatures [38]. This unusual temperature dependence of plasticity has been attributed to complex secondary relaxation spectra and shear band dynamics in metallic glasses [33–38]. In contrary, Maass et al. reported that higher plasticity associated with multiple shear bands at low temperature is an artifact resulting from inhomogeneous stress development at the sample-anvil interface during compression testing [39]. It remains unclear why such an artifact will have a temperature dependence reported in cryogenic studies of metallic glasses.

Effect of testing temperature on shear band propagation in metallic glasses has been directly measured from the displacement rate at different temperatures [35]. Substantial reduction in shear band speed with decreasing temperature has been reported. The effect of temperature on shear band nucleation is inferred largely from fracture analysis, calorimetric measurements, and secondary

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relaxation mechanisms [35,38,40]. Higher shear band density observed in metallic glasses fractured at low temperatures is attributed to enhanced nucleation [33,35,37,40]. This correlation is however inconclusive because the samples tested at different temperatures fracture at varying strains and shear band nucleation is a sequential process. Ideally, the samples deformed to a fixed strain value at different temperatures should be compared to understand the effect of temperature on nucleation of shear bands. Such controlled experiments are difficult in uniaxial compression due to irreproducibility of shear bands formed in such tests [41]. In contrast, bending creates controllable shear bands which are suitable to study the effect of temperature on shear band characteristics (density, spacing, critical length etc.). Here, we use bending of Pt-based ($\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$) and Zr-based ($\text{Zr}_{35}\text{Ti}_{30}\text{Cu}_{8.25}\text{Be}_{26.75}$) metallic glasses from liquid nitrogen to glass transition temperatures to investigate the effect of temperature on shear bands and the plasticity.

2. Experimental

Pt-based metallic glass was synthesized by water quenching technique described elsewhere [42]. Zr-based metallic glass was acquired from LiquidMetal technologies. Amorphous state of both metallic glasses was verified using DSC (differential scanning calorimeter) and XRD (x-ray diffraction). The glass transition temperature (T_g), crystallization temperature (T_x), and Poisson ratio (ν) for Pt-based and Zr-based metallic glasses are listed in Table 1. The Poisson ratio was calculated from the transverse and longitudinal velocity values measured using ultrasonic technique. Rectangular plates were prepared by thermoplastic forming followed by water quenching to ensure similar thermal history. Thickness (t) of plates was 0.7 mm and 1.5 mm for Zr-based and Pt-based samples, respectively. The thicker samples for Pt-based metallic glass were used to achieve higher fracture strain in our bending setup. The samples were bent around the mandrels of varying radii (r). The bending strain ($\epsilon = t/2r$) was calculated from the mandrel radii and the sample thickness as described in the work of Conner et al. [7]. We used a custom-built mandrel setup which can be heated or cooled to vary the testing temperature (Fig. 1). Sub-zero temperatures were obtained using dry ice and liquid nitrogen while high temperatures were achieved by resistive heating cartridges. Temperature close to the test sample was measured using two thermocouples. The bending time was shorter than the relaxation and the crystallization times at every testing temperature. Therefore, it can be assumed that the as-quenched glassy state was retained during mechanical testing. No visible discoloration due to oxidation was observed after high temperature testing.

3. Results and discussion

Fig. 2 shows the fracture strain of Pt-based and Zr-based metallic glasses tested at different temperatures. The fracture strain values are an average of three bend tests conducted at every temperature. At temperatures above $\sim 0.8T_g$, the metallic glasses deformed homogeneously through viscous flow to accommodate large strains without fracture. Below $0.8T_g$, the samples deformed in a shear

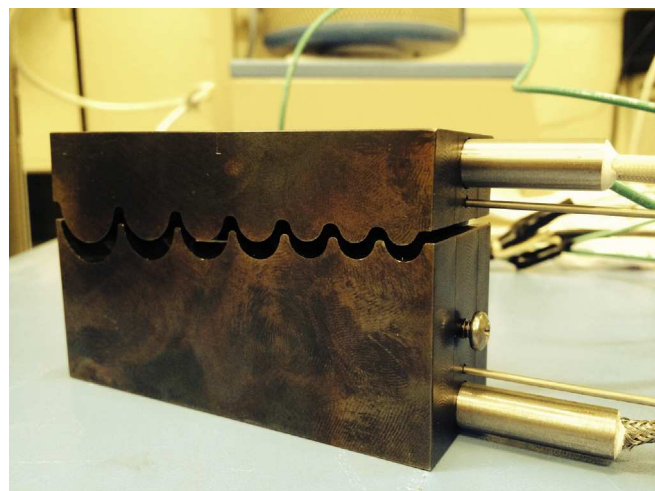


Fig. 1. Custom-built bending setup used in present experiments. The testing temperatures from 77 K to 550 K were achieved using liquid nitrogen and high temperature heating elements.

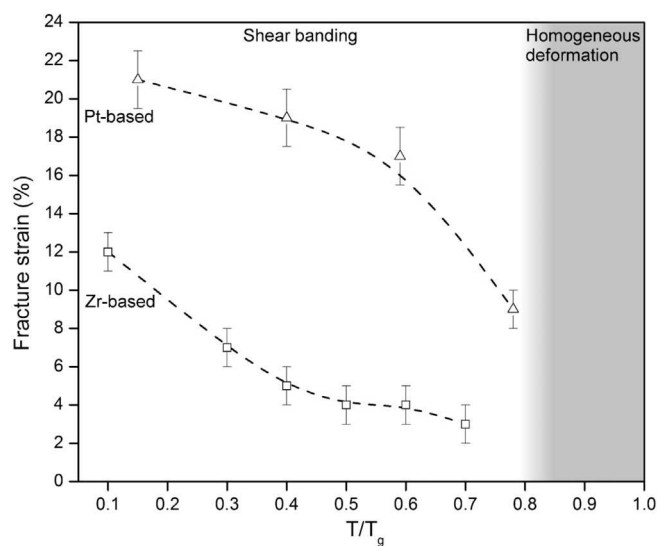


Fig. 2. Temperature dependent fracture strain of Zr-based and Pt-based metallic glasses measured at temperatures from 77 K to $0.8T_g$. Both metallic glasses show decrease in fracture strain with increasing temperature. The data in homogeneous flow regime are not shown because the samples did not fracture.

localized manner and the fracture strain decreased with increasing temperature. The fracture strain decreased from 21% to 9% for the Pt-based metallic glass with increasing temperature from 77 K ($0.15T_g$) to 393 K ($0.78T_g$), respectively. Similar temperature dependence can be observed for the Zr-based metallic glass though its fracture strain is lower than Pt-based metallic glass due to different elastic constants. Metallic glasses with higher Poisson ratio or lower shear modulus to bulk modulus ratio tend to be more ductile [4]. Increase in bending plasticity with decreasing temperature observed here is consistent with several previous studies on other metallic glass formers tested in compression [33–35].

Overall plasticity of metallic glasses is controlled by the nucleation and propagation of shear bands before one of them becomes an unstable crack [1]. To understand the correlation between the temperature dependent plasticity and shear band activity, the fractured samples were analyzed using SEM (scanning electron

Table 1

Glass transition temperature (T_g), onset of crystallization temperature (T_x), and Poisson ratio (ν) of Pt-based and Zr-based metallic glasses.

Metallic glass	Composition (at.%)	T_g (K)	T_x (K)	ν
Pt-based	$\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$	489	572	0.41
Zr-based	$\text{Zr}_{35}\text{Ti}_{30}\text{Cu}_{8.25}\text{Be}_{26.75}$	611	719	0.37

microscopy). Fig. 3a shows SEM images of tensile side of Zr-based metallic glass samples fractured at various temperatures. Noticeable changes observed in shear band features due to temperature variation are quantified in Fig. 3b. Shear band spacing (λ) increased or equivalently the shear band density decreased with increasing temperature. However, the propagation length (l) of shear bands at fracture became shorter at higher temperature. The critical shear offset which is proportional to the propagation length at fracture is an indicator of intrinsic plasticity of metallic glasses [7, 43]. Therefore, reduction in shear band length suggests that the metallic glasses become less resistant to crack formation at higher temperature. As discussed later, these results cannot be explained by the temperature dependence of elastic constants alone.

Enhanced plasticity of metallic glasses at cryogenic temperatures has been explained by increased nucleation of shear bands [33–37]. The number of shear bands decrease with temperature (Fig. 3a) but it cannot be unambiguously stated if this is a cause or consequence of temperature dependent plasticity. Because the samples fractured at different strains are expected to display different shear band densities due to successive nucleation of shear

bands during plastic deformation. To study the effect of testing temperature on shear band nucleation, constant strain tests were performed at varying temperatures. Pt-based metallic glass samples were bent to 7% strain without fracture at 77 K, 298 K, and 393 K. The SEM micrographs clearly reveal distinct shear band characteristics to accommodate the same strain at different temperatures (Fig. 4). At 77 K, more shear bands of shorter lengths are formed whereas fewer and longer shear bands are observed with rising temperature. In addition, the number of secondary shear bands also decreased at elevated temperatures. These results indicate that lower temperature promotes shear band nucleation as the other parameters (sample size, applied plastic strain, and strain-rate) were kept constant. Similar results were obtained for the Zr-based metallic glass samples bent to 5% strain.

Our bending experiments on Zr-based and Pt-based metallic glasses reveal two important findings: (1) the shear band density for any applied strain decreases and (2) the propagation length (or critical shear offset) at fracture becomes shorter with increasing temperature. These two effects determine the overall plasticity of metallic glasses in the shear localized regime. Here, we evaluate

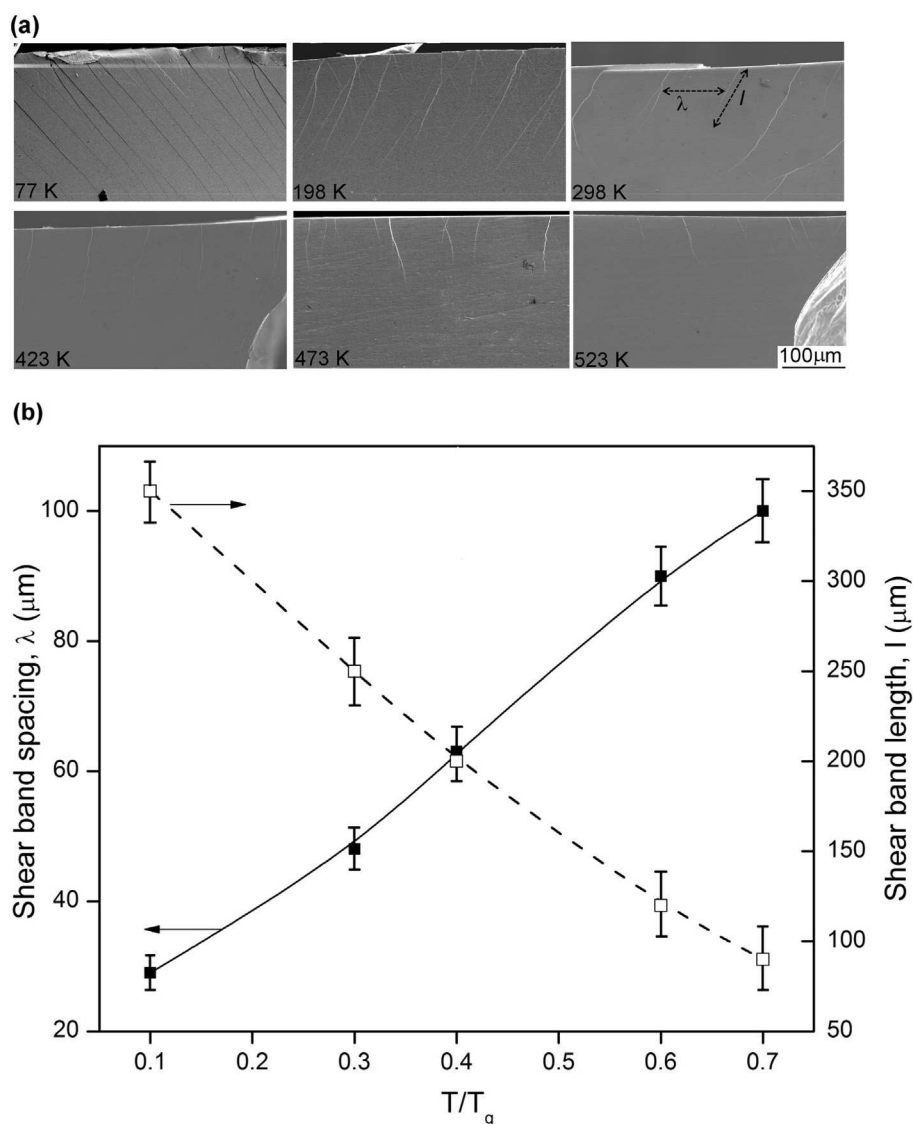


Fig. 3. SEM micrographs of tensile side of Zr-based metallic glass samples fractured at various temperatures (a). The shear band spacing and the propagation length measured from the SEM images reveal strong temperature effects on shear banding process (b).

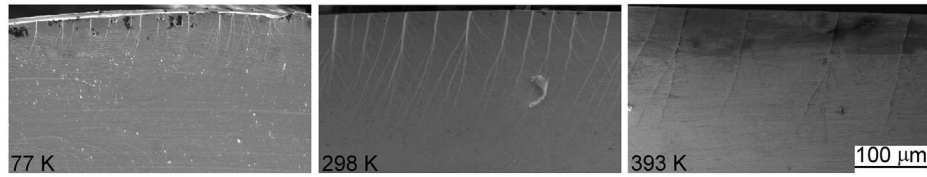


Fig. 4. SEM micrographs of tensile side of Pt-based metallic glass samples bent to 7% strain at 77 K, 298 K, and 393 K. Fewer and longer shear bands are formed to accommodate the same strain at elevated temperatures.

theoretical aspects of shear band nucleation and propagation to rationalize the temperature dependence. Plastic deformation in metallic glasses is governed by rearrangement of clusters of atoms referred to as shear-transformation-zones (STZs) [2,44]. Based on the Argon's STZ model, Schuh et al. proposed a three stage mechanism for shear band formation from the collective dynamics of STZs [44,45]. In the first stage, spatially uncorrelated STZs form at weak sites such as an excess free volume or chemical heterogeneities. These STZs result in a local strain field and structural disordering, which bias the formation of subsequent STZs in the vicinity. The localized collections of STZs formed in second stage can be considered as shear band nuclei. When the strain-rate mismatch between these nuclei and the surrounding matrix diverges towards infinity, rapidly propagating shear bands are formed (third stage). The quantitative analysis revealed that the formation of viable shear band nuclei from STZs (second stage), is temperature dependent. Schuh et al. derived an expression for the strain-rate ratio ($\dot{\Gamma}_o$) in the shear band nucleus to that of the surrounding glass as [45]:

$$\dot{\Gamma}_o = \frac{\dot{\gamma}_n}{\dot{\gamma}} \propto \exp\left(\frac{C}{T}\right), \quad (1)$$

where $\dot{\gamma}_n$ is the strain-rate in the shear band nuclei, $\dot{\gamma}$ is the strain-rate in the surrounding matrix, and C is a constant that combines the Boltzmann constant and the elastic energy stored in the shear band nuclei as a result of an STZ operation [45]. The $\dot{\Gamma}_o$ determines the viability of an embryonic nucleus to transform into a fully propagating shear band. The nucleus with higher $\dot{\Gamma}_o$ value requires lesser strain to nucleate a shear band. According to Eq. (1), lower testing temperature will promote nucleation of shear bands by increasing the early stage localization $\dot{\Gamma}_o$. Therefore, many STZ clusters can become strain bearing shear bands at low temperatures leading to higher shear band density. This can explain our constant strain results where lower testing temperature resulted in multiple shear bands of shorter lengths. However, the change in critical shear band length with temperature cannot be explained by the nucleation model.

Critical shear band length (or shear offset) beyond which it evolves into a crack, is measure of fracture toughness which scales with Poisson ratio for metallic glasses [43]. Weak but positive temperature coefficient for Poisson ratio has been reported for La-based and Ni-based metallic glasses [40,46]. Such temperature dependence of Poisson ratio would predict an increase in critical shear band length for metallic glasses at high temperatures which is in contrary to the experimental findings. Therefore, temperature dependence of elastic constants does not explain the temperature effects on plasticity of metallic glasses. Demetriou et al. combined the elastic constants and the activation barriers for shear flow and cavitation to describe the crack initiation in a shear band [43]:

$$\log(f) \sim \frac{T_g}{T} \left(\frac{B}{G} - 1 \right), \quad (2)$$

where B and G are the shear and the bulk modulus, respectively. The dimensionless parameter f measures the capacity for shear flow before cavitation in an operating shear band. Although, Eq. (2) was originally used to compare the fracture toughness of different metallic glass formers at ambient conditions, it can accurately describe the temperature dependence, not captured by the elastic constants. According to Eq. (2), f decreases with increasing temperature because B/G (or Poisson ratio) is a weak function of temperature. Therefore, cavitation in a shear band is thermally assisted, leading to earlier fracture at elevated temperatures.

A similar conclusion can be drawn from the hypothesis of liquid-like layer formation during shear banding. It has been proposed that a liquid-like layer develops ahead of propagating shear band due to temperature rise. A shear band results in catastrophic failure when the size or the viscosity of the liquid-like layer reaches a critical value [47–49]. This mechanism is supported by the cross-sectional SEM analysis of samples fractured at different temperatures (Fig. 5). All the fractured surfaces show a smooth region which corresponds to the shear offset caused by propagation of a shear band. The smooth region is followed by typical *vein* morphology for liquid fracture. Therefore, the smooth region can be considered as the shear band propagation length required for formation of liquid-like layer viable for crack initiation and extension. It is clear from the SEM images that the smooth region decreases with increasing temperature in Pt-based (Fig. 5a) and Zr-based (Fig. 5b) metallic glasses. These results suggest that higher temperature facilitates shear band to crack transformation in metallic glasses. Miracle et al. reached a similar conclusion based on theoretical analysis of shear band as a planar source of heat [48]. They showed that the size of liquid-like layer depends inversely with ΔT^2 (where $\Delta T = T_g - T$). This model was used to explain the brittleness of low- T_g metallic glasses such as, Ce-based, Mg-based, and La-based [48]. It can also be used to analyze the effect of varying temperature on the fracture behavior. Increase in testing temperature (or decrease in ΔT) will result in formation of critical sized liquid-like layer at a smaller shear band sliding.

Thus, liquid-like layer hypothesis and the cavitation model (Eq. (2)) both predict decrease in the extent of plastic deformation in a shear band before crack opening at higher temperature. This along with reduced nucleation of shear bands can explain the diminishing plasticity of metallic glasses with increasing temperature. It is worth noting that we did not observe a peak in plastic strain at temperatures around $0.4T_g$ as reported in a recent work [38]. Our experiments show continuously increasing plasticity up to 77 K, the lowest temperature investigated here. However, decrease in plasticity at temperatures lower than 77 K cannot be ruled out from the present work.

4. Conclusions

In summary, bending plasticity and shear band characteristics of Zr-based and Pt-based metallic glasses were studied at cryogenic

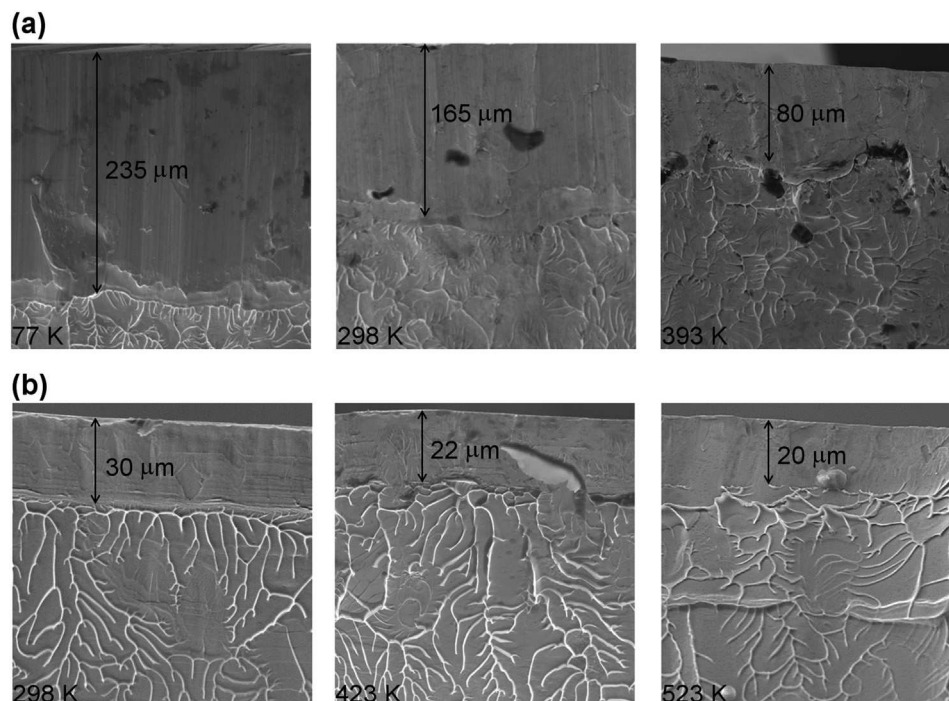


Fig. 5. Cross-sectional views of Pt-based (a) and Zr-based (b) metallic glass samples fractured at different temperatures. All fractured surfaces exhibit two distinct morphologies on the tensile side (smooth region and veins). The size of smooth region decreases with increasing temperature indicating an early failure at elevated temperatures.

and elevated temperatures. Plasticity decreased with increasing temperature which is a consequence of reduced shear band nucleation and critical propagation length. Increase in testing temperature affects the shear band nucleation by reducing the early stage localization of STZs. Temperature also promotes the shear band to crack transformation resulting in early failure at elevated temperatures. These temperature effects on shear bands are captured by the nucleation model and the cavitation or liquid-like layer hypotheses for crack opening.

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