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Single shear-band plasticity in a bulk metallic glass at cryogenic temperatures

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At cryogenic temperatures bulk metallic glasses can sustain higher plastic strains than at room temperature. This is generally believed to result from an intrinsic shear-band nucleation rate that increases with decreasing temperature. Here we report on inhomogeneous flow operating via a single shear band even at cryogenic temperatures, challenging the presupposition of increased shear-band activity. The results provide a new interpretation of non-serrated flow and explain, via a simple viscosity law, the correspondingly observed strength increase with decreasing temperature.

Keywords: Metallic glass; Plasticity; Low temperature; Shear band

Bulk metallic glasses (BMGs) belong to a class of materials that do not contain crystalline order and therefore do not admit dislocation structure and activity as seen in their crystalline counterparts—the fundamental microscopic mechanism of plasticity that underpins our understanding of materials strength and toughness of experimentally realizable metals in relation to the theoretical strength of a perfect metallic crystal. Despite the strong atomic-scale disorder of BMGs, these materials exhibit a high elastic limit and high fracture strength, good fracture toughness, soft-magnetic properties, and superplastic formability near the glass transition [1–4]. However, because of their lack of a dislocation network, they are unable to strain harden or to be cold-worked at temperatures well below their glass transition temperature T_g, which has so far hampered our ability to understand the underlying physics of their plastic deformation.

A major hurdle hindering room-temperature engineering applications of BMGs is their pronounced brittleness under tensile loading and their limited compressive ductility, which at low homologous temperatures (T/T_g) is carried by highly localized flow defects, called shear bands, which are confined to a thickness of only a few tens of nanometers [5,6]. A tremendous international effort is underway to improve the room-temperature ductility of BMGs, both via intrinsically modifying the glassy structure or by processing composite structures [7–9]. The resulting studies have shown that the increase in shear-band density is the dominating factor for increasing the ductility, because the applied plastic strain can be distributed over a large number of shear bands, i.e. a larger fraction of the sample volume [10,11]. Further, the spread in strain at failure in compression testing at ambient conditions can be correlated with the boundary conditions of the test, showing that non-uniform compressive deformation at room temperature yields a higher shear-band density and consequently a larger strain prior to failure [12,13].

At cryogenic temperatures, however, a clear picture of enhanced strength and ductility at a stable stress level has been reported with decreasing testing temperature [14–19]. A testing series as a function of decreasing temperature generally comprises a strain-rate- and temperature-dependent transition from serrated flow at room temperature to smooth, non-serrated flow at sub-ambient temperatures [20–23]. More specifically, this transition is linked to the shear-band velocity at the chosen testing temperature [24]. When passing from room temperature to cryogenic temperatures, a consistent increase in shear-band density has been reported both in tension and compression, and this also provides the basis for rationalizing the increased ductility [14–16,18,25]. The phenomenon of a higher shear-band density at lower temperatures has been widely attributed to a reduction

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of (i) the mobility of free volume and (ii) the propagation rate of the shear bands, both yielding an increase in the number of sites of shear-band nucleation [5,15,16,18,19,26]. Given that low-temperature deformation is intrinsically related to a higher shear-band nucleation rate, and thus shear-band density, resulting in a greater ductility, BMGs have been classed as highly promising materials for cryogenic applications. Due to the proposed increased shear-band nucleation rate with decreasing temperature, it has hence been concluded that deformation in the non-serrated regime (low temperature) should not occur on a single shear band.

In this paper we uncover the opposite, i.e. the BMG specimens deform on one shear band only, even at cryogenic temperatures but with immediate strain softening occurring upon yielding. Our results allow us to develop a new interpretation of non-serrated flow in terms of shear-band nucleation rate and shear-band propagation. Using this approach we are also able to explain the significant strength increase of BMGs with decreasing temperature.

Uniaxial compression tests were conducted using cylindrical samples of glassy $Zr_{52.2}Ti_5Cu_{17.9}Ni_{14.6}Al_{10}$ (Vit105) specimens of 3 mm in diameter with an aspect ratio of 5/3. The samples were prepared by suction casting in an arc-melter, and carefully polished to ensure plane-parallel ends. Displacement-controlled tests were conducted in the temperature range of 77–298 K and at a strain rate of $10^{-3}\ s^{-1}$. An extensometer bridging the upper and lower compression platens was used to record displacement, and a piezoelectric load cell was employed to measure the load during the tests. Scanning electron microscopy (SEM, Hitachi SU-70) was used to image the surface morphologies.

Figure 1 displays three successive stress–time curves for a specimen tested at 213 K, where the reload was shifted in time such that a continuous curve is formed. Once unloaded, the sample was immediately reloaded. For instance, at the end of load 1, the drive direction of the cross-head was reversed at an arbitrary moment after flow had begun until close to zero force was measured. Without delay the subsequent reloads were conducted in a similar fashion. Each loading cycle exhibits a transient yield phase with a stress overshoot before the stress decays in a smooth manner. Such stress overshoots are known from the homogeneous flow of BMGs, and are typically associated with a non-Newtonian flow regime, where the internal structural relaxation processes are not fast enough to keep up with the external loading rate [27]. Each loading cycle with the transient stress overshoot followed by non-serrated flow corresponds to the initiation of shear-band propagation, where shear-band arrest remains absent due to the high applied strain rate relative to the shear-band velocity at that temperature [28]. Since the stress overshoot magnitude increases with each reload from 13 to 49 MPa, eventually reaching 80 MPa, it may be concluded that the relative degree of structural relaxation increases as a function of accumulated damage created during each of the three unloading/loading cycles. This result thus directly demonstrates shear-band structure relaxation, leading to a shear-initiation stress upon each reloading that exceeds the previous unloading stress.

In addition to this unexpected flow response, SEM investigations reveal that the sample deformed via only a single shear band that was reactivated during each reloading cycle (see inset of Fig. 1). The fact that only a single shear band was activated places non-serrated flow, regarded as a flow regime with a shear-band nucleation controlled mechanism [5,14–16], in a new light. The observation that non-serrated flow can occur with a single flowing shear band does not agree with the view that the applied strain rate cannot be relieved quickly enough into the surrounding matrix leading to multiple shear-banding events and thus a high shear-band nucleation rate, producing a higher shear-band density. From the presented data it becomes clear that all strains can be imparted into a single shear band at a forced rate. Another remarkable difference between what is generally found in the literature on cryogenic plastic flow of BMGs and the curve seen in Figure 1 is the absence of a stable stress level with increasing deformation.

The demonstrated flow response shown in Figure 1 was also reproduced at 158 and 233 K. The present work thus demonstrates that an understanding of inhomogeneous deformation in BMGs at cryogenic temperatures is contained in the physics of a single flowing shear band. However, due to the material's high sensitivity to the testing boundary conditions [12,13], only a few samples showed this flow response, because of multiple shear-band initiation at the sample–anvil interface. In order to improve the number of samples deforming on a single shear band, defined 50 µm root radius notches were introduced 1 mm below the top surface; this applies in the remainder of this document. These notches serve as first shear-band initiation sites only, whereas subsequent flow again underlies the evolving test dynamics, as in the un-notched case.

To further investigate the flow response of a single shear band at cryogenic temperatures, additional tests were conducted. Compression curves obtained at 123 K (curve I), 173 K (curves II–IV) and 298 K (curve V) are summarized in Figure 2. Of particular interest is the direct comparison of stress–strain curves obtained at the same strain rate ($10^{-3}\ s^{-1}$) and temperature (173 K) stemming from (a) the conventional case of a dense shear-band morphology (curve IV, as-cast specimen

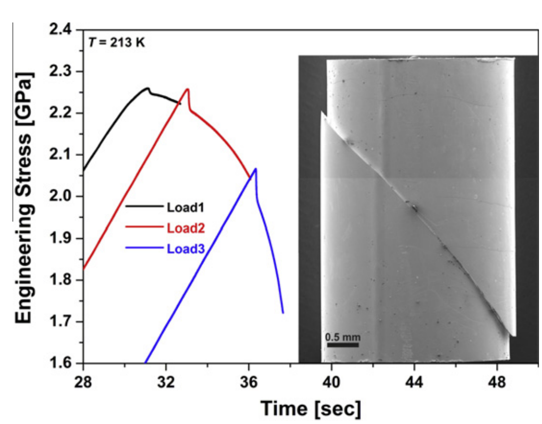

Figure 1. Three successive loading cycles of a specimen tested in the non-serrated regime at 213 K. The inset demonstrates that the sample deformed via a single shear band.

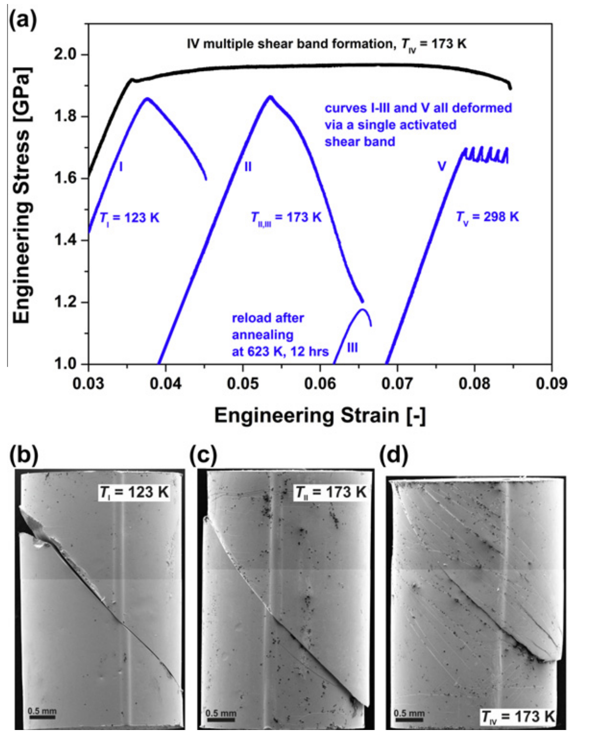

Figure 2. (a) Stress–strain curves obtained at 123 K (curve I), 173 K (curves II–IV) and 298 K (curve V). Curves I–III and V are from samples that flowed in a single shear band. A direct comparison is displayed via the different flow behaviours in a sample that exhibits stable plastic flow (IV, as-cast specimen geometry), high ductility and a high shear-band density, and a sample that flows via a single shear band (II) accompanied with a rapid stress decay once yielding occurs. A flow curve of a sample tested in the serrated regime at 298 K is also displayed (curve V). The lower images (b–d) show SEM micrographs of the samples after testing, related to curves I, II and IV in (a).

geometry), and (b) a specimen that flowed on a single shear band (curve II). The difference in stress decay rate is very pronounced and can be directly linked to the evolving shear-band structure, as demonstrated in Figure 2c and d.

Figure 2d shows clearly that the sample exhibiting stable plastic flow developed a dense shear-band structure, whereas the two samples to the left (Fig. 2b and c) only contain a single shear band for which flow exhibits a rapidly decaying stress level (curves I (123 K) and II (173 K): see Fig. 2a). A large fraction of the shear bands in Figure 2d exit at the anvil–sample interface, this being a typical signature of a successive nucleation of shear bands due to geometry-constrained plasticity that maintains a stable flow stress [12,13]. It can thus be concluded that the often-reported stable and extensive plastic flow in the non-serrated regime is a result of an inhomogeneous stress state resulting from mechanical constraints rather than from a specific intrinsic materials property enhancing the shear-band nucleation rate and thus flow at cryogenic conditions. The intrinsic flow behaviour of the metallic glass is reflected by the activation of a single shear band, which rapidly suffers from flow softening once the peak stress is reached. Note that the dramatic flow softening of curve II in Figure 2 is only to a small extent due to the reducing cross-sectional surface, which amounts to 1.5%, whereas structural flow-softening processes must be responsible for the remaining stress decay of 98.5% or 645 MPa over a plastic strain of 1.2%.

Figure 1 demonstrated stress overshoots upon reloading. These are attributed to structural relaxation that reduces the flow defects in the formed shear band during unloading, leading to a higher yield stress at reloading. In order to explore the possibility of rejuvenation of the shear-band structure, the sample displayed in Figure 2c (flow curve II in Fig. 2a) was subjected to an annealing treatment at 623 K (50 K below T_g) for 12 h in an argon atmosphere. This annealing treatment is not sufficient to cause mechanical embrittlement [24]. The sample was subsequently passed to a second loading cycle (Fig. 2a, curve III) under conditions identical to the first loading (curve II in Fig. 2a). It is clear that the yield stress of the annealed specimen hardly reaches the unloading stress of the as-cast sample. SEM investigations revealed that the failed sample indeed reactivated the same single shear band as during the first loading cycle (not shown here). This result can be understood in the framework of nanovoid formation and microcracking that has been reported to develop within the shear band with increasing strain localization prior to crack initiation [29]. Both nanovoids and microcracking are irreversible structural effects that need to be considered in order to explain the non-measurable increase in yield stress after the annealing treatment.

Despite the very different flow response compared to earlier work on cryogenic flow and the clear dependence of the flow stress on the evolution of the shear-band morphology, the increase in yield stress (here unambiguously defined as the maximum stress appearing at the elastic–plastic transition) as a function of decreasing temperature is preserved (Fig. 3). The data obtained here is compared to literature values of the maximum stress [15,30] and the yield stress [17] of various metallic glass alloys as a function of temperature. Although different measures of stress were used, it is apparent that the temperature dependence of the strength is similar.

The temperature dependence of the macroscopic yield strength is therefore not dependent on the evolving

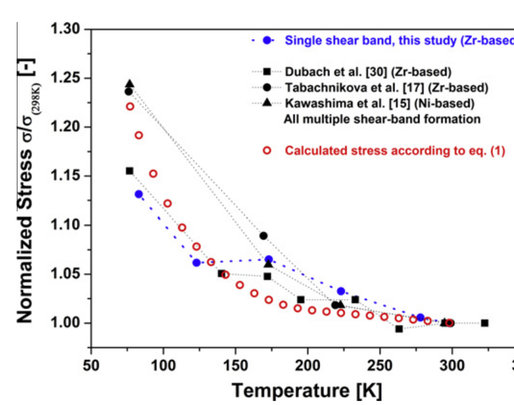

Figure 3. Normalized stresses as a function of temperature from this study and literature data [15,17,30]. The graph shows that the relative increase of the yield stress or maximum attained flow stress with decreasing temperature can be explained by a rate-dependent model (calculated stresses according to Eq. (1)), and that it is not affected by the different shear-band morphologies.

shear-band morphology, whereas any other measure of stress, including geometrically induced strain hardening, does depend on it. Current literature links the strength increase to a decrease in effective atomic distance and a resulting increase in elastic stiffness at cryogenic temperatures [15,18,31]. The reported increase in elastic modulus at 77 K is, however, only between 1% and 5%, which is not sufficient to explain a strength increase of up to ~25%. This suggests a different dominating link between yield stress and strain at yield than the elastic constants [32]. The origin of this increase in flow stress is thought to lie in rate-induced effects due to imposing an applied strain rate in the non-serrated regime. That means we assume a rate-dependent increase in flow stress after passing the transition from serrated to non-serrated flow, because hereafter the shear band experiences effectively an ever-increasing imposed rate when distanced from the transition condition between serrated and non-serrated flow. Thus the underlying assumption is in analogy to a rate-dependent flow stress of non-Newtonian media once non-serrated flow is entered. For any strain rate and temperature above the transition ($\dot{\varepsilon}_{\mathrm{appl}} > \dot{\varepsilon}_{\mathrm{trans}}$), ($T < T_{\mathrm{crit}}$) and $m(T < T_{\mathrm{crit}}) > 0$, the yield stress may be approximated by an equivalent flow stress, i.e.

$$\sigma(T,\ \dot{\varepsilon}_{\mathrm{appl}}) = \sigma_0(T)\left[\frac{\dot{\varepsilon}_{\mathrm{appl}}}{\dot{\varepsilon}_{\mathrm{trans}}(T)}\right]^{m(T)} \tag{1}$$

where m is the strain-rate sensitivity and σ_0 is a reference stress whose temperature dependence is assumed to be dominated by changes in elastic modulus only (see above). With input data from Refs. [21,28,30] for $m(T)$ and $\dot{\varepsilon}_{\mathrm{trans}}$, Eq. (1) can be evaluated for an applied strain rate of $10^{-3}\ \mathrm{s}^{-1}$ and a corresponding transition temperature T_{crit} of approximately 200 K. Above 200 K, the yield stress is simply taken to be equal to the reference stress $\sigma_0(T)$, which is assumed to increase linearly by 3% from room temperature down to 77 K. The normalized stress results obtained by Eq. (1) are plotted along with the experimental data in Figure 3, showing that the rate-dependent model can indeed explain the marked increase in stress below the transition temperature.

In conclusion we provide experimental results that allow for a new interpretation of non-serrated flow in BMGs, where it is demonstrated that inhomogeneous flow at cryogenic temperatures can be entirely understood by the physics of a single flowing shear band—a result that does not require an increased shear-band nucleation rate with decreasing temperature. Moreover, we show that single shear-band activity can exhibit close to unconstrained plastic flow accommodating all imposed strain at the applied rate via significant strain softening. Thus the enhanced stable plasticity seen during non-serrated flow at cryogenic temperatures is a result of an evolving shear-band density due to geometrical constraints imposed by the deformation experiment, and is not an intrinsic property of the BMG. The observed stress–temperature scaling is independent of shear-band morphology, and can be successfully modelled by non-Newtonian behaviour respecting the transition strain rate between serrated and non-serrated flow. Reactivation of a single shear band displays stress overshoots, offering a promising means to investigate the relaxation dynamics of individual shear bands. This novel route is expected to provide further fundamental insight into the physical behaviour of a shear band, and thus into the fundamentals of inhomogeneous deformation of BMGs or disordered materials in general.

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