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Comparative analysis of inhomogeneous plastic flow in bulk and ribbon metallic glasses monitored by acoustic emission

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ABSTRACT

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1. Introduction

The strongly spatially localised and temporally inhomogeneous deformation is a feature of the mechanical response of metallic glasses at low homologous temperature, where they show only very limited ductility. The low ductility of metallic glasses is one of the main factors distracting these materials from broader possible applications. It has been commonly understood that with the control over localisation processes - that is possible with the modern glassy alloys making – one can enhance the ductility, e.g. [1]. This challenge has lead to a surge in research activity in this area over past two decades. However, the understanding of the physical origin of localisation mechanisms remains rudimentary up to date owing to many experimental and theoretical uncertainties in characterisation of amorphous structures. The most popular theoretical views describing the strain localisation in metallic glasses in association with softening and viscosity reduction in shear bands rely heavily on the concepts of "free volume" [2,3] and shear transformation zones (STZs) [4]. Despite their elegance and simplicity, none of these can accurately describe the nucleation and propagation of shear bands in metallic glasses. The applicability of these concepts to the homogeneous and/or inhomogeneous plastic flow has been questioned in the literature [5,6] and many of the parameters in the current theories cannot unambiguously be related to the thermomechanical history of the glass [7]. However, several well-established characteristics of the inhomogeneous plastic

Aiming to gain a better insight into the phenomenological description and mechanisms of inhomogeneous plastic deformation in metallic glasses, we review the currently available experimental data on the acoustic emission (AE) behaviour in these materials under load and compare the results for typical ribbon-shaped and bulk metallic glasses. It is shown that the AE patterns reflecting the kinetics of shear banding are very much alike for both kinds of glasses (as-cast and as-spun) despite a gross difference in the quenching rate during manufacturing, geometry and loading conditions. It is also shown that the stress drops during serrated plastic flow in BMGs are triggered by short time localised shear events having a comparatively large scale although no simple correlation between the stress drops and AE amplitudes is found.

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flow are worthy noticing: plastic strains tend to localise in narrow shear bands of 10–20 nm width and up to several micrometers step offset. The strain rate in the shear bands reaches extremely high value as of $10^9 \, \text{s}^{-1}$ [8]. In addition it has been recently recognised that serrated plastic flow is intimately related to single shear steps seen on compression test samples, which have magnitudes of several hundred nanometers to few micrometers [9,10]. With regard to the deformation kinetics, improvement in the understanding has been gained through the evaluation of strain rate sensitivity, which below a critical temperature changes from negative to positive values [11,12]. Despite these new insights into shear band mechanisms in metallic glasses it is still a matter of question how to describe the nucleation and mutual propagation of single atomic shear events to develop into macroscopic shear bands.

Acoustic emission (AE) is a phenomenon which accompanies rapid local structural rearrangements in solids, providing thereby an intuitive insight into the kinetics and dynamics of shear banding at the macroscopic phenomenological level in a real time scale. In the present brief communication, we review and compare the currently available experimental data on the AE behaviour during straining of typical ribbon [13,14] and bulk [15] metallic glasses, where the necessary conditions of locality and rapidness of structural rearrangements are evidently met in the shear bands (see the previous paragraph). Although some apparent similarity between AE in ribbon-shaped and bulk metallic glasses (BMG) has already been noticed in [16], the detailed comparison of the AE behaviour in two kinds of glasses has not been performed. Furthermore, no features associated with the serrated flow in BMG were noticed and discussed in [16] since the testes were performed in tension and

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Fig. 1. Loading curves and AE time history in terms of peak amplitude for (a) a typical ribbon-shaped MG tested in tension and (b) a typical BMG tested in compression; nominal strain rate in both cases is 1×10^{-3} s⁻¹.



Fig. 2. AE amplitude distributions obtained for a ribbon-shaped MG tested in tension (a) and a BMG tested in compression (b); both distributions correspond to the time histories presented in Fig. 1.

cyclic ramp loading. Since the general AE appearance is similar in different metallic glasses [12–15] regardless of the precise chemical composition, we have chosen typical amorphous metallic ribbons and bulk samples for illustrative purposes.

2. Experimental

The ingots of $Zr_{65}Ni_{10}Cu_5Al_{7.5}Pd_{12.5}$ alloy (composition is given in nominal atomic percentages) were prepared by arc-melting in an argon atmosphere.¹ Bulk rod samples of 3 mm × 3 mm cross-section were prepared by the copper-mold injection and gravity casting techniques. Ribbon-shaped $Fe_{75}Co_5B_{10}$ metallic glasses of 30 μ m thick and 10 mm width were obtained by a standard rapid quenching melt-spinning technique. The samples were examined by conventional X-ray diffractometry with monochromatic Cu K α radiation: no traces of crystallinity were found.

A mechanically polished 3 mm \times 3 mm \times 6 mm BMG specimen for compression testing was set between two parallel ceramic platens. The miniature low-noise broadband AE sensor MSAE-1300WB with a built-in 27 dB preamplifier and a 30 kHz high-pass cut-off filter has been securely mounted directly on a lower platen and axially centred with the specimen. Vacuum oil was used as a coupling media to ensure a good acoustic contact between the transducer and the platen.

The ribbon-shaped glasses were tested in tension using the same screw-driven machine. All tests have been performed at the nominal strain rate of 10^{-3} s⁻¹.

The signal from the output of the preamplifier was transferred through a low-noise filter-amplifier MSAE-FA-010 and then acquired via the AE-recording system triggered by threshold-crossing and operating at 6.25 MHz sampling rate,

12 bits amplitude resolution and 8k samples recorded per single realisation. The details of AE signal processing involving the statistical and spectral analysis of the AE time-series in application to metallic glasses have been reported elsewhere [17,18].

3. Results and discussion

A typical AE record in terms of the peak amplitudes is shown in Fig. 1. Although virtually no macroscopic plasticity is noticed in the ribbon-shaped glass, AE starts at fairly low stresses, Fig. 1(a), and exhibits a long history before fracture. A similar behavour is seen for the BMG where AE commences at stresses much lower than the macroscopic yield stress. AE events reflecting rapid shear events in metallic glasses are irreversible either in ribbon [13,14,17-19] or bulk [16,20] samples. Evidently, AE appears a random time-series of temporarily well-separated bursts having different amplitudes. Corresponding amplitude distributions are plotted in Fig. 2. The most common feature of these distributions is their bimodalilty [13], which indicates that two types of AE sources having significantly different average amplitudes may operate in metallic glasses under load (tensile or compressive). The AE amplitude is fundamentally associated with AE source scale and velocity as proposed in a simplified mechanistic model by Scruby et al. [21]. For example, for a dislocation loop expanding on the slip plane at the velocity v to a final radius a in a semi-infinite body, a maximum displacement (which is commonly supposed to be proportional to the AE amplitude if the transducer is mounted at the epicenter) at the surface

¹ The specimens were prepared in the Institute of Materials Research, Tohoku University.



Fig. 3. AE bursts typical of ribbon MG (a and b) and BMG (c and d), corresponding to low-amplitude fraction (a and c) and high-amplitude fraction (b and d) of the distributions shown in Fig. 2.

at a distance r to the source is given as

$$U_z = \frac{c_T^2}{c_L^3} \frac{bav}{r} \tag{1}$$

where c_L and c_T are the longitudinal and the shear wave velocities, respectively.

Typical waveforms of AE transients corresponding to low- and high-amplitude fractions of the distribution histogram are shown in Fig. 3. Again, the similarity between the waveforms of the signals belonging to the same class (low- or high-amplitude) is evident. It has long been recognised that the shear bands serve as primary sources of AE in metallic glasses. Clear evidence for that was obtained in the *in situ* experiments in the SEM chamber [13]. Later it was also shown that the amplitude of AE transients correlates with the length of the shear band [16] in line with Eq. (1).

The similarity between the AE parameters in both the as-cast and as-spun metallic glasses extends further if one uses a sort of data squeezing procedures including factor analysis and signal categorisation techniques such as those used in [17,18]. Results of the so-called *k*-means clustering are shown in Fig. 4: AE signals fall naturally into two categories (clusters) with centroids which are far apart in the selected coordinates: AE amplitude vs. the median frequency of the AE power spectral density. The results of cluster analysis agree nicely with the simplistic amplitude analysis, Fig. 2, suggesting the presence of two kinds of sources. Supplementary discussion on the analogies of the statistical properties of AE time-series in the as-cast and as-spun samples can be found in [16].

Turning to the features of the serrated flow, one should notice the following: (i) high-amplitude (energy) AE events align with the onset of every yield drop, Fig. 5(a). (ii) Low-amplitude AE bursts are randomly scattered between the stress drops, Fig. 5(b), serving as precursors for larger AE events as has been discussed in [8] for ribbon-shaped metallic glasses, where the relation between the low- and high-amplitude AE events was investigated using linear location technique. Besides, no apparent correlation is found between the magnitude of the stress drop and the AE amplitude, (Fig. 6).

Hence, the conclusion, which can be plausibly drawn from the comparison of the AE behaviour in ribbon and bulk metallic glasses, is that the elementary mechanisms controlling the inhomogeneous



Fig. 4. AE signal clustering in f_m – U_p space for a ribbon-shaped (a) and BMG (b).



Fig. 5. AE transients corresponding to the yield drops in the Zr-based BMG sample tested in compression: (a) high-amplitude AE corresponding to the yield drops and (b) low-amplitude AE randomly distributed during the stress rise between the successive serrations.

plastic flow (with or without serrations in the loading curve) in metallic glasses are essentially the same regardless of the specimen geometry and type of loading. However, one should bear in mind that Zr-based BMGs belong to a group of materials having a high glassy forming ability and therefore they can be produced at quenching rates which are about four orders of magnitude smaller than those attained in melt-spinning techniques employed commonly in manufacturing of ribbon-shaped metallic glasses. Using positron annihilation, it has been proven that an increase of the quenching rate by three orders of magnitude results in a nearly linear increase of the annihilation time, being indicative of a proportional increase of a "frozen in" free volume in a glass [22]. Comparison of the AE behaviour during strain localisation in typical metallic glasses obtained at significantly different quenching rates, shows that most propounded features of shear banding kinetics and dynamics are strikingly the same, regardless of the thermal history during manufacturing and the geometry of the specimens. This indicates that the underlying microscopic mechanisms of shear band formation and propagation in metallic glasses are also similar and are not strongly dependent of the quenching rate in fair agreement with [8] where it is argued that the basic shear mechanism in all metallic glasses is the same. This is also supported by an ample similarity between the morphology of shear bands and fracture surface of ribbon-shaped and BMGs, as has been reported repeatedly. Jiang et al. [23] have compared the mechanical behaviour of the as-cast bulk and as-spun ribbon Cu₆₀Zr₃₀Ti₁₀ metallic glasses and showed that both the bulk and ribbon alloys exhibit a similar serrated plastic flow during the nanoindentation test. Very similar specific relief patterns around the indents were also observed in both alloys. The excess free volume is generally regarded as a principal structural factor affecting the mechanical response of metallic glasses [8]. Of course, there is no doubt that the overall mechanical behaviour and ductility is affected by manufacturing [24] and the following thermal history of a metallic glass. However, the above results rise up a question whether or not the free volume plays a key role in the inhomogeneous plastic flow of metallic glasses. As a matter of discussion, the issue, which has yet to be addressed, is related to multiple shear banding and the go-and-stop intermittent character of the shear band propagation: if the strain localisation is caused by the catastrophic softening due to stress-induced free volume production (e.g. [3]), why the fracture does not occur immediately along the very first shear band? The stress dropping, which is seen during the serrated flow in compression, cannot answer this question simply because many shear bands are seen in tension whereas the stress drops are not observed [12,16-18]. The selfsupporting avalanche-like aggregation of the free volume, resulting in the drastic softening, should lead to inevitable fracture along the



Fig. 6. AE amplitude vs. stress drop magnitude during the serrated plastic flow of the $Zr_{65}Ni_{10}Cu_5Al_{7.5}Pd_{12.5}$ bulk glassy alloy tested in compression with the strain rate of $1 \times 10^{-3} \text{ s}^{-1}$. No apparent correlation is noticed.

first shear band. The AE investigations show convincingly that this is not the case and plastic shearing is blocked many times before fracture. Specific mechanisms involving nanocrystallisation within the shear band, which can be responsible for local hardening and slip blockage, are not assumed here as they do not represent a general case. To resolve this controversy, a free-volume-exhaustion mechanism was proposed in a qualitatively manner by Yang et al. [25] to explain the multiple shear banding in metallic glasses. However, this mechanism has yet to be justified both experimentally and theoretically.

In conclusion, we have reviewed (although still qualitatively) the behaviour of AE in metallic glasses and compared it for the typical as-cast Zr-based BMG and as-spun ribbon Fe-based glassy alloy. Of course, the comparison should be performed more systematically and quantitatively using one and the same alloy produced with different quenching rates. Nonetheless, the present superficial observations revel that there is an ample similarity between the shear bands kinetics in bulk and ribbon-shaped metallic glasses.

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