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Understanding the serrated flow and Johari-Goldstein relaxation of metallic glasses

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ABSTRACT

An increasing tendency of serrated flow with increasing loading rate of a La-based (La) metallic glass (MG) with pronounced Johari-Goldstein relaxation in nanoindentation is investigated, which is abnormal compared to precedent observations where the serrated flow normally decays with increasing loading rate as discovered in the referenced Zr-based (Zr) and Pd-based (Pd) MGs. Based on systematic analysis, a unified dimensionless number is proposed to elucidate the different origins of the abnormal serrated flow of the La MG and the normal serrated flow of the Zr and Pd MGs. The abnormal serrated flow behavior of the La MG provides critical implications in understanding the role of Johari-Goldstein relaxation in the flow of metallic glasses.

1. Introduction

The origin of the plasticity of metallic glasses [1] (MGs) has been explored with great efforts by studying the rich deformation phenomena of metallic glasses under various circumstances, from monolithic to compound [2], from static to dynamic [3], and from macroscopic to microscopic [4]. Shear band [5] consisting of self-organized shear transformation zones (STZs) [6,7] dominates the plastic deformation of metallic glasses, except in creep [8] or in the deformation of nanometer-sized samples [4]. Due to the "prone-to-failure" character of shear bands, global plasticity of metallic glasses is available only under constrained deformation modes, such as indentation [9], compression [10], and bend [11] etc. In these cases, a distinct trait of the load-displacement curve is the "zigzag" periodical discontinuity of plastic deformation in time domain, i.e. intermittently "burst-out" plastic events, termed serration or serrated flow [12]. Therefore, the mechanism of serrated flow is of great importance to understanding the origin of the plasticity of metallic glasses.

Serrated flow has been widely observed in a variety of metallic glass systems both in physical experiments [13] and in atomistic simulations [14]. The observed serration is proposed to correspond to either the activation of an embryonic shear band or the elementary propagation event of a mature shear band [15]. The statistical character of the size of serrations is related to the nonlinear activation dynamics of the elementary plasticity events in the form of chaos state or self-organized critical state which suggests different ductility of metallic glasses [16]. The stick-slip behavior is also adopted to characterize the serrated flow of metallic glasses based on the liquid-like to solid-like transition inside shear bands [17]. In reference to the Portevin–Le Châtelier (PLC) effect in crystal alloys, the serration in the negative strain rate sensitivity regime is explained similar to the dynamic strain aging effect in the deformation of solid solutions [18]. Regardless of the above different explanations, a widely accepted fact is that the serrated flow is strain rate dependent as high rates would usually reduce the intensity of serration [19].

To evaluate the shear band dominated serrated flow of metallic glasses, nanoindentation [20] provides an effective method for its extraordinary high resolution in space on nanometer-sized scale which coincides with the thickness of shear bands of metallic glasses [21]. Moreover, the "constrained" deformation mode in indentation enables versatile and flexible examination on the plastic deformation of metallic glasses in the avoidance of shear banding fracture. On the load (P)-displacement (h) curve in nanoindentation tests, the serrated flow of metallic glasses often takes a staircase-like form and presents as "pop-in" events where the displacement exhibits a "forward-jump" at an almost constant load [19]. Normally, the size of the "pop-in" events in nanoindentation would decrease with increasing loading rate at room temperature [22]. Nevertheless, in this work, we report an abnormal, i.e., loading rate-enhanced, serrated flow of a La-based (La) MG [23] with pronounced Johari-Goldstein relaxation [24] and provide a self-consistent explanation to this unusual phenomenon by *in-situ* examining the creep behavior of metallic glasses under the indenter with the displacement-time curve during the load-hold stage of the indentation program. A unified dimensionless number is proposed to elucidate the different origins of the abnormal serrated flow of the La MG and the normal serrated flow which decreases with increasing loading rates. More importantly, whilst that the Johari-Goldstein relaxation

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has been related to the activation of STZ in the flow of metallic glasses [25], its nature, especially the underlying atomic dynamics involved, remains subtle and elusive [26]. The current work on the abnormal serrated flow of La MG with pronounced Johari-Goldstein relaxation provides meaningful information for identifying the role of Johari-Goldstein relaxation in the flow of metallic glasses.

2. Experimental

Alloy sheets of a size of 2 mm × 30 mm × 40 mm are prepared by copper mould casting under Ti-gettered Ar atmosphere from master alloys of the typical nominal composition: Zr ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$, Vit1), La ($La_{62}Al_{14}Ag_{2.34}Ni_{10.83}Co_{10.83}$), and Pd ($Pd_{40}Cu_{30}Ni_{10}P_{20}$). The amorphous structure of the cast alloy sheets is examined by X-ray Diffraction (XRD) and differential scanning calorimeter (DSC). The samples for nanoindentation were polished to mirror finish before experiments. Nanoindentation tests are conducted on an MTS Nano Indenter XP™ with a Berkovich diamond tip. Load control mode and a group of loading rates $\dot{P}$: 0.33mN/s, 1.32mN/s, 13.2mN/s, and 70mN/s are selected. The indentation program is composed by the following stages: (1) load to a maximum force P_{max} of 200mN at a constant $\dot{P}$; (2) maintain the load for 10 s at the maximum force P_{max}; (3) unload to 10% of the peak load at a rate of 10mN/s; (4) hold the load for 10 s to perform thermal drift calibration; (5) unload completely. To guarantee the reliability of the load (P)–displacement (h) curve, each test is repeated 7 times.

3. Results

Fig. 1 shows the P-h curves of the La ($La_{62}Al_{14}Ag_{2.34}Ni_{10.83}Co_{10.83}$), Zr ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$, Vit1), and Pd ($Pd_{40}Cu_{30}Ni_{10}P_{20}$) MGs in nanoindentation. For a clear view, the curves are shifted along the transverse coordinates. The P-h curve obeys the power law equation [27]: $P = Ch^m$, where C and m are fitting constants depending on loading rate and the shape of indenter. By fitting the P-h curves with the power law equation, it is found that m is about 1.8 ~ 2 for all the 3 MGs, regardless of the different indentation depths and loading rates, implying similar flow behavior of the 3 MGs on the continuum scale. In Fig. 1, it can be seen that prominent "pop-in" events can be found on the P-h curves of Pd MG (Fig. 1c). However, compared to that of Pd MG, the Zr MG (Fig. 1b) shows slight serrations and the La MG (Fig. 1a) shows no obvious "pop-in" events in the selected loading rate range of the present work.

For a better view on the serrations [28], the P-h curves are subtracted by their fitted curves with the power law equation: $P = Ch^m$. As shown in Fig. 2, the Δh-P curve, i.e. Δh at the same load P, is the difference between each P-h curve and its power law fit. Different serrations can be discerned on the subtracted P-h curves of all the 3 MGs at different loading rate $\dot{P}$. Interestingly, it can be seen in Fig. 2a that with increasing loading rate $\dot{P}$, distinct serrations with a lower frequency than that of the Zr (Fig. 2b) and Pd (Fig. 2c) MGs emerge on the subtracted P-h curves of La MG. This is abnormal in contrast to the serrations of the Zr and Pd MGs which decreases with increasing loading rate $\dot{P}$. Similar abnormal serrations have also been found in other LaAl(Cu, Ni) MGs [28] and Ce-based MGs [29] but at lower loading rates around 0.03mN/s, where a low glass transition temperature (T_g) is the main cause of the abnormal serrated flow. In previous works, it is proposed that the low T_g makes the room temperature (298 K) a relatively high reduced temperature (0.7 ~ 0.8T_g) at which the viscous flow of metallic glasses is possible at loading rates below 0.03mN/s. The abnormal serrated flow phenomenon is attributed to $\dot{P}$ induced transition from viscous flow to shear band dominated deformation. However, for the La MG in this work, the T_g is 450 K, which gives a reduced value of 0.67T_g for room temperature at which the nanoindentation tests are conducted. This value is below the critical value of 0.7T_g proposed in the literature [9,28] where locates a crossover from decreasing serrated flow

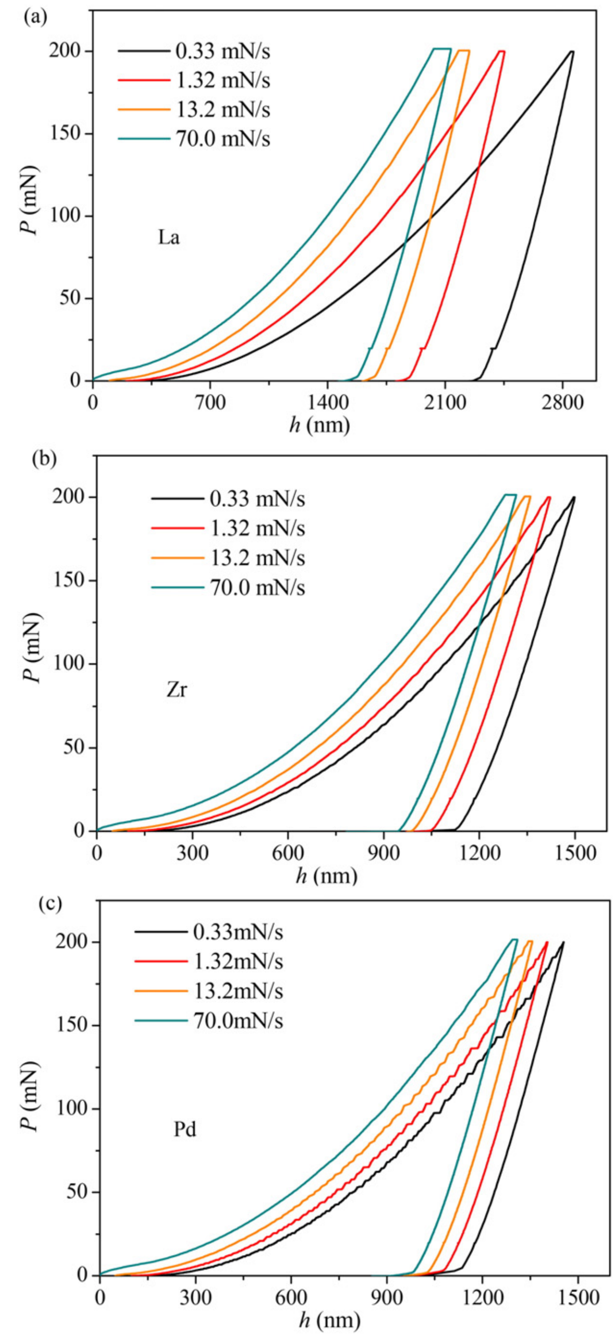

Fig. 1. Load-displacement P-h curves of La (a), Zr (b), and Pd (c) metallic glasses. The curves are shifted along the transverse coordinates.

with increasing $\dot{P}$ to increasing serrated flow with increasing $\dot{P}$. More importantly, according to the flow regimes of metallic glasses [9], for the relatively high loading rates adopted here, the transition from viscous flow to shear band dominated deformation might not be the dominant reason for the abnormal serrated flow of the La MG in Fig. 2a.

To pursue the dominant reason, the *in-situ* deformation state of metallic glasses at different loading rates under the indenter would be highly useful. Fortunately, the load-hold stage of the indentation program actually functions as a "deformation frozen" technique and captures the *in-situ* creep deformation behavior of metallic glasses under the indenter [30]. The load-held period of 10s is also in favor of the elimination of the deviations of the indentation depth caused by thermal drift which would grow pronouncedly with extended holding time