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Dynamic behavior of a Ce-Al bulk metallic glass

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Abstract. The mechanisms of stress relaxation in metallic glasses under high strain rates are an area of active study. The lack of extended structure forces strain accommodation through alternative modes to slip. For example, amorphous Ce_3Al has been shown to undergo a phase transition to the crystalline FCC Ce_3Al at 25 GPa under quasistatic loading. Whether this mechanism extends to high strain rates has yet to be determined. We present results of an initial study into the ultrafast deformation characteristics of a Ce-Al bulk metallic glass. Using the Janus laser at the Jupiter Laser Facility (LLNL), thin targets 30 micron in thickness were shocked over a range of pressures up to 30 GPa. The velocity of the target rear surface was measured using a line-imaging VISAR to reveal features in the wave profile attributed to stress relaxation. In addition, experiments were performed on crystalline forms of Ce-Al prepared through heat treatment of the amorphous material. Preliminary results reveal a distinct precursor wave above and below 1.5 GPa, which gives way to a complex multiwave structure around 1.5 GPa, most likely indicative of a polyamorphic transition.

1. Introduction

Bulk metallic glasses (BMGs) are solids composed of a combination of metallic elements often quenched from melt to suppress the formation of a crystalline lattice. This lack of structure leads to interesting physical behavior under dynamic compression, with regards to both yield phenomena and high pressure phase transformations. One area of active study is on the microscale mechanisms of stress relaxation in metallic glasses under high strain rates. Previous work has shown the lack of extended structure and regular atomic coordination in metallic glasses to frustrate conventional modes of strain accommodation through plastic slip, leading often to rapid failure along discrete bands of thermoplastic instability [1]. BMGs also exhibit rich phase transition behavior, with the potential to undergo both polyamorphic and crystallization transitions under compression [2-5].

The Ce-Al glass system provides an opportunity to study this diverse range of behaviour. Through static high pressure studies, Ce_3Al metallic glass has been shown to undergo a polyamorphic transition from a low density to high density amorphous phase between 1.5-5 GPa, characterized by an 8.6% volume collapse arising from delocalisation of the 4f electrons in Ce [4, 5]. At pressures in excess of 25 GPa, the amorphous alloy has been observed to crystallise into a single FCC crystal [6]. Whether these transitions take place at high strain-rates has yet to be determined, as there have been very few studies on the dynamic response of these materials [7].


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In this work, we describe a preliminary study of the ultrafast deformation behaviour of an amorphous Ce_3Al alloy subjected to laser-driven shock loading, up to pressures exceeding the crystallisation threshold established from earlier static high pressure studies.

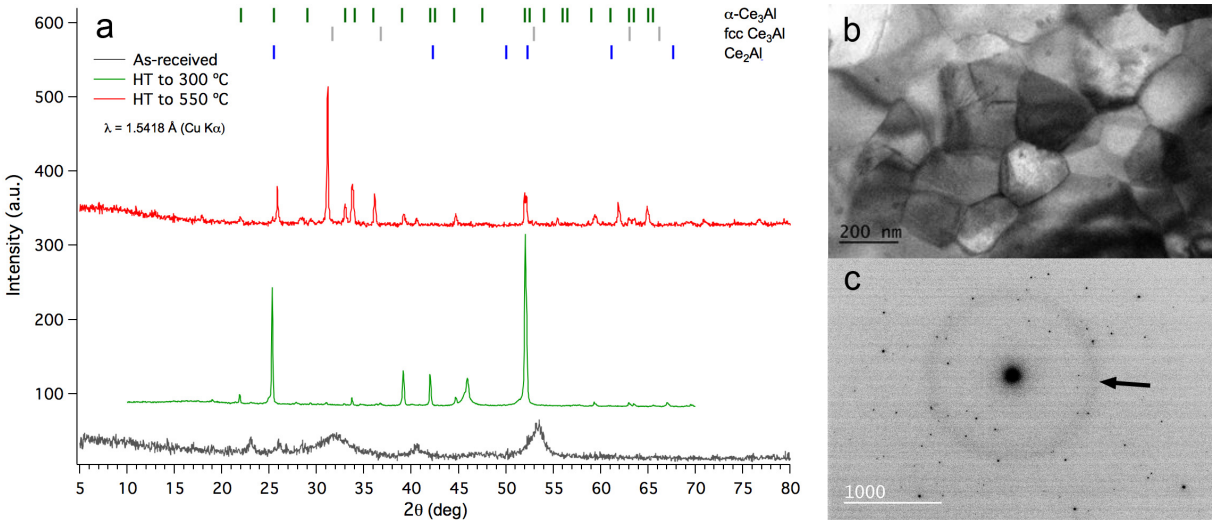

Figure 1. (a) X-ray diffraction scans for the as-received Ce_3Al ribbons and samples heat treated to 300 and 550 °C. The broadened features in the as-received ribbons reveal possible crystallinity on the nanoscale, but otherwise a lack of long-range ordering. Upon heating the samples crystallise into a complex combination of phases. (b) TEM of the sample heat-treated to 300 °C, showing grains on order 200 nm in size. (c) An electron diffraction pattern from the region in (b) which shows a faint ring indicating the presence of residual amorphous material.

2. Materials and Characterisation

Amorphous ribbons of Ce_3Al, approximately 30 μm thick and 2 mm wide, were fabricated through melt spinning by Ames Laboratory. A batch of the ribbons was heat treated at 550 °C for 1 hr in order to induce crystallization and yield a stoichiometrically-equivalent metal standard with which to compare behaviour of the amorphous material. The as-received and heat-treated material was characterised using X-ray diffraction, the results of which are shown in figure 1a. The as-received, amorphous material exhibits broad, undefined peaks indicative of a material lacking extended crystallinity; however, although the diffraction peaks are very broad, they are still somewhat pronounced in certain areas, indicating that there exist nanocrystallites within the amorphous microstructure. In contrast, the samples heat-treated to 300 and 550 °C exhibit clear diffraction peaks, consistent with an extended crystalline phase. For comparison, the uppermost markers in the figure correspond to the diffraction lines of several Ce-Al phases; though the pattern clearly contains a mixture of several phases, best agreement is obtained with the hexagonal-structured α-Ce_3Al phase.

The extent of crystallinity was further studied through TEM of the material heat-treated to 300 °C. The bright field image shown in figure 1b reveals a microstructure composed of many grains on the order of 200 nm in size. However, the electron diffraction pattern taken over the same region, shown in figure 1c, reveals that although the sample is predominantly crystalline, there is residual amorphous material, as indicated by the diffuse amorphous ring.

3. Experimental Method

The Ce_3Al ribbons were shocked over a range of pressures up to 30 GPa using the Janus laser at the Jupiter Laser Facility (LLNL). The targets were affixed to support washers and irradiated with a high-energy laser pulse operating at 527 nm, and delivering anywhere between 15-100 J over 8 ns, driving a supersonic pressure wave in the material. The velocity of the target rear surface was measured using a line-imaging VISAR to reveal features in the wave profile attributable to stress relaxation or phase transformation. Additionally, a thin sheet of acrylic was placed behind the target to facilitate post-shock recovery, tilted at 8° to eliminate back reflections. The configuration of the target, recovery reservoir, drive beam and VISAR diagnostic are shown in figure 2a, with a representative raw VISAR fringe record shown in figure 2b.

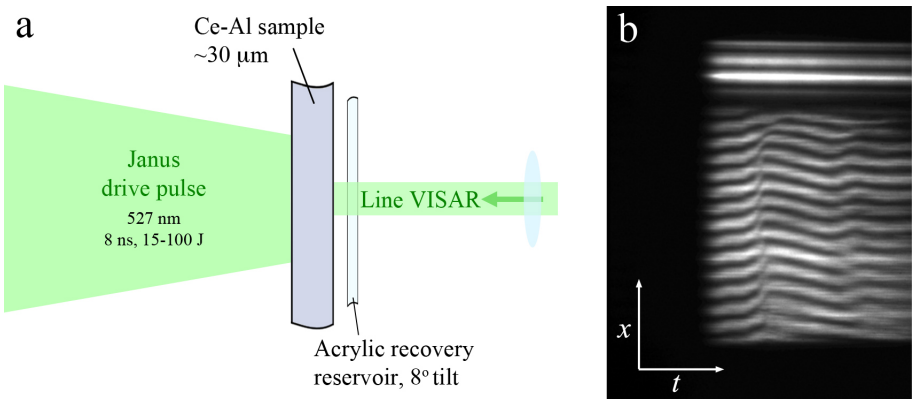

Figure 2. (a) Schematic showing the target and diagnostic configuration in the laser shock experiments. (b) A representative line VISAR record for the amorphous ribbon, with space and time increasing along the indicated directions.

4. Results and Discussion

The amorphous Ce_3Al was compared to heat treated Ce_3Al at similar drive conditions. It can be seen in figure 3a that the initial low-amplitude wave, referred to here as the precursor, is of higher amplitude and broader extent in the amorphous material than in the heat treated sample. Additionally, the magnitude of the pullback feature in the amorphous sample, indicative of the spall strength of the material, is nearly double the same in the heat treated sample despite being at a lower drive energy. As shown in the raw line VISAR record of figure 3b, the heat treated sample exhibited rapid loss of reflectivity after shock breakout. This loss of reflectivity combined with the lower apparent spall strength could be the result of failure and surface breakup.

Next, a comparison was made between the behaviour of amorphous samples driven over a range of increasing pressures. Figure 4 plots three VISAR lineouts from experiments loading amorphous targets to 1 GPa, 2.3 GPa and 3.3 GPa. The intermediate pressure, at 2.3 GPa, experiences a decrease in particle velocity that is not seen in the 1 GPa or 3.3 GPa cases. As mentioned before, Ce_3Al experiences a transition from low density amorphous (LDA) to high density amorphous (HDA) between 1.5-5 GPa, resulting in an 8.6% volume reduction [4]. The occurrence of this polyamorphic transition under dyanamic loading could be the cause of the shoulder at about 200 m/s in the VISAR for the line-out from the sample shocked to 2.3 GPa.

To explore this hypothesis further, the degree of compression during loading was approximated by converting the particle velocity profiles for the 1.3 and 2.3 GPa cases into

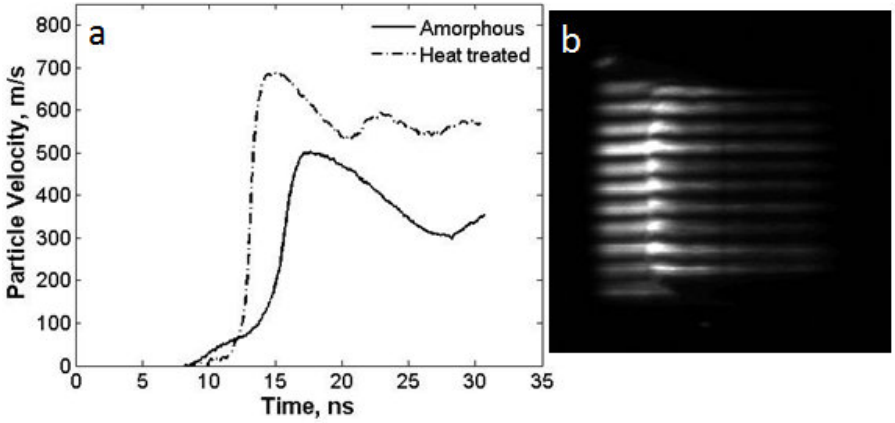

Figure 3. (a) A comparison between the particle velocity profiles of the amorphous and heat-treated Ce_3Al ribbons, driven with 25 and 28 J, respectively. The amorphous target displayed a broad, low-amplitude precursor, a common feature observed in disordered materials. In contrast, the precursor in the heat-treated target was ∼80 % smaller and led abruptly to the primary wave front. (b) A raw line VISAR record for the heat-treated Ce_3Al sample shows rapid loss of reflectivity upon shock break-out, which may be due to surface break-up.

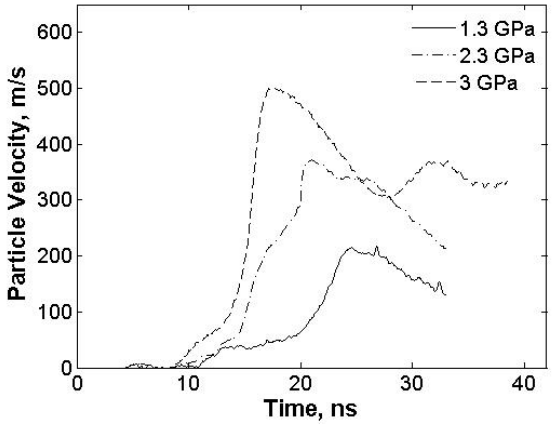

Figure 4. Particle velocity profiles measured for the amorphous target at increasing drive energies; the traces are labelled according to the calculated stress at the peak state. In each trace, the broad precursor shoulder is clearly observed, leading to a single primary loading wave for the lower stress condition. For the sample driven to 2.3 GPa, an intermediate loading wave is observed, which appears to become over-driven by 3 GPa.

stress and strain using the following differential equations,

$$\delta\epsilon = \delta u_p / C_L \tag{1}$$

$$\delta\sigma = \rho_0 C_L^2 \delta\epsilon \tag{2}$$

where $\delta\epsilon$ is infinitesimal change in strain, δu_p is infinitesimal change in particle velocity, C_L is longitudinal wave speed, $\delta\sigma$ is infinitesimal change in stress, and ρ is initial density [8]. It can be seen in figure 5 that the second peak from the 2.3 GPa line-out in figure 4 corresponds to a

drop in pressure change and a reduction in volume of about 7% . This is not dissimilar to the degree of collapse associated with the polyamorphic transition seen in the quasi-static work since data is indicative of a transition which slows the particle velocity and it occurs at approximately the same pressure with approximately the same volume reduction. The values are not entirely matched but this is likely due to the simple wave approximation used to produce the normal stress vs volume relationship. Investigation of the other shots at higher stress states did not show such a trend indicating a phase transition; most likely, the samples were over-driven. TEM will be also conducted on recovered samples to identify any retained phases.

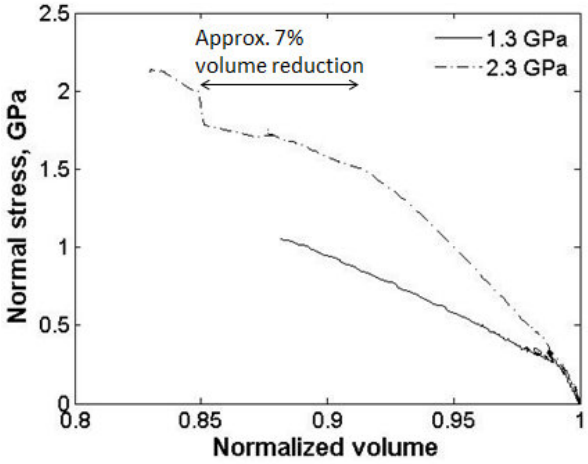

Figure 5. Stress vs. specific volume for the amorphous Ce_3Al shocked to 1.3 and 2.3 GPa. An approximately 7% volume collapse is observed at 1.5 GPa, which is of similar magnitude to the LDA-HDA transition previously observed by Zeng et al. [4].

5. Conclusions and Further Work

The dynamic response of a Ce-Al metallic glass was investigated using laser-driven shock loading while running interferometric velocimetry on the rear surface to determine the particle velocity and infer the stress-volume states achieved. Measurements revealed distinct differences between the deformation behaviour of the amorphous and crystalline alloy. The possible signature of the LDA - HDA transition was observed at 1.5 GPa under dynamic loading. The crystallization transition was not observed in the VISAR but recovery analysis using a TEM will help to determine if one occurred at all.

Future work will try to capture the crystallization transition during dynamic compression by running the VISAR streak camera at higher temporal resolution and performing in-situ diffraction. Static experiments will include diffraction while heating amorphous Ce-Al to characterize the temperature dependency on microstructure.

Acknowledgments

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