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Flaw tolerance vs. performance: A tradeoff in metallic glass cellular structures

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Abstract

Stochastic cellular structures are prevalent in nature and engineering materials alike. They are difficult to manipulate and study systematically and almost always contain imperfections. To design and characterize various degrees of imperfections in perfect periodic, stochastic and natural cellular structures, we fabricate a broad range of metallic glass cellular structures from perfectly periodic to highly stochastic by using a novel artificial microstructure approach based on thermoplastic replication of metallic glasses. For these cellular structures, precisely controlled imperfections are implemented and their effects on the mechanical response are evaluated. It is found that the mechanical performance of the periodic structures is generally superior to that of the stochastic structures. However, the stochastic structures experience a much higher tolerance to flaws than the periodic structure, especially in the plastic regime. The different flaw tolerance is explained by the stress distribution within the various structures, which leads to an overall "strain-hardening" behavior of the stochastic structure compared to a "strain-softening" behavior in the periodic structure. Our findings reveal how structure, "strain-hardening" and flaw tolerance are microscopically related in structural materials.
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1. Introduction

Imperfections are almost inevitably present in materials, and they can impose dramatic effects on the materials' properties. In many cases, imperfections even overshadow the intrinsic properties of materials. Examples can be found in all major material classes. In ceramics, imperfections in the form of pores and inclusions dramatically degrade mechanical properties [1]. Imperfections in the form of controlled impurities play a key role in semiconductors and are utilized to improve electrical [2], optical [3] and magnetic [4] performances. In crystalline metals, defects in the form of dislocations practically control plastic behavior. An in-depth understanding of dislocations has therefore been the focus of the metallurgical community over the last 60 years. A material class that is prevalent in engineering applications and in nature is cellular materials. For these broadly used materials, the effects of imperfections on performance have been scarcely studied [5–7]. Formally, cellular materials can be categorized into periodic and stochastic (non-periodic) structures, both of which almost always contain some imperfections. The majority of current fabrication methods limit the control of imperfections and hence prevent a systematic flaw tolerance study. Such a limitation has motivated computational simulation studies of defect behavior for both periodic and stochastic structures [8–16]. However, due to the limited predictability of constitutive equations in describing the plastic flow as well as the difficulty in

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simulating non-linear behavior such as post-buckling and cell walls, reasonable agreements have only been achieved for the elastic interactions mechanical behavior [8–14]. For a fundamental understanding of the effects of imperfections on the mechanical response of cellular structures, including their plastic behavior such as plastic flow and energy absorption capacity, a systematic study on precisely fabricated structures with precisely implemented imperfections encompassing a broad range of structures and materials is required.

The performance of cellular materials is governed by their cell geometries, relative density and by the material used within. A broad and systematic study requires the consideration of a variety of both materials and geometries. A material class that covers a wide range of behaviors is bulk metallic glasses (BMGs). The absence of crystalline grains, dislocations and grain boundaries in the amorphous structure results in a homogeneous material with near-theoretical strength and high elasticity [17–20]. The length-scale-dependent plasticity in BMGs, i.e., the smaller the length, the larger their ductility [21–26], has also been explored in overall micro-architecture design [27–32]. Furthermore, mechanical properties of BMGs are highly dependent on the configuration they resume, which can be explained within the energy landscape concept [33,34]. Dramatic changes in ductile or brittle behavior can be achieved via processing, such as changing the cooling rate [35] or structural relaxation [35–38]. To cover a wide range of materials, we use BMGs in both its as-formed and relaxed state. Specifically, we use $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ (LMX) BMG in the as-formed state, which combines high strength, modulus and ductility [19]. Through structural relaxation, a precisely controllable change in the BMG's energetic state, its properties can be altered to high strength, high modulus and low ductility (brittle) [36]. We also use a typical thermoplastic polymer, polyether ether ketone (PEEK), as a representative material with low strength and low modulus combined with high ductility. Through a processing opportunity, so-called thermoplastic forming (TPF) [39], we show how to precisely fabricate a vast variety of BMG structures consisting of different cell geometries covering a wide range of relative densities, including those which contain precisely controllable imperfections. We focus on the most common imperfections, a missing, imperfect or fractured cell wall ligament, which we assume by a missing wall. Considering periodic and stochastic structures made of materials ranging from strong to weak, and from brittle to tough, allows for a broad and systematic study revealing the general behavior of how imperfections affect mechanical properties of cellular materials.

2. Experimental

2.1. Sample preparation

Here, we consider LMX BMG as the parent material in both its ductile, as-formed state and also its brittle state, achieved through structural relaxation. As a representative of a low strength, low modulus material, we use commercial standard PEEK (purchased from McMaster-Carr). Master alloy ingots of $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ were prepared by arc-melting a mixture of the pure elements in an argon atmosphere. The amorphous state was achieved by water quenching in a 10 mm diameter quartz tube. The amorphous nature of the BMGs was confirmed by X-ray diffraction (XRD-6000 Shimadzu) and differential scanning calorimetry (Perkin Elmer Diamond DSC).

For periodic cellular structures, we consider the representative case, hexagonal honeycombs, which are by far the most common type. Delaunay tessellation is an attractive prototype for stochastic structures because of its simplicity and similarity to engineering and natural cellular structures [40,41]. Here, by using Delaunay tessellation similar to that described in Refs. [42,43], two stochastic structures, stochastic 1 (S1) and stochastic 2 (S2), with different structure factors were constructed, the latter of which is more disordered than the former (Appendix A).

The existence of a supercooled liquid state in BMG formers gives rise to TPF, a processing method where BMGs can be formed under similar conditions as thermoplastics [39,44,45]. TPF-based processing has been developed over length scales ranging from meters to angstroms [39,44,45]. It often allows for the fabrication of geometries that were previously unachievable with any other metal processing methods [46]. We employ TPF-based molding to fabricate the various cellular structures considered here by replicating them from Si molds. Within our fabrication method, we first imported the coordinate information of the various cellular structures into AutoCAD software to construct a photomask with a Heidelberg DWL-66 laser beam writer. Photolithography and deep reactive ion etching were then performed to fabricate Si cellular structure molds with an etching depth of $\sim$400 μm. Replication of the cellular structures was achieved by TPF of the BMG into the Si molds at 420 °C under a pressure of $\sim$20 MPa, after which extra BMG was carefully removed by grinding while the Si molds were etched with 20% KOH solution (Fig. 1). For PEEK samples, a pressure of $\sim$2 MPa at $T = 340$ °C was used. In order to examine the influence of defects on the mechanical properties of cellular structures, various numbers (4, 8 and 18) of cell walls were randomly removed from the structures (with a total number of 350 cell walls) in the AutoCAD layout design and the resulting molds were replicated with the various materials considered here. The cellular structures considered in this work include: perfect periodic hexagonal structure, periodic structures with 4 ($\sim$1%), 8 ($\sim$2%) and 18 ($\sim$5%) walls missing, and S1, S2 stochastic structures, S1 and S2 structures with 4 ($\sim$1%), 8 ($\sim$2%) and 18 ($\sim$5%) walls missing. For each structure with a specific number of missing walls, three different locations of the missing wall sites were considered to achieve an average assessment. It should be mentioned that we chose locations for the multiple defects that were spatially decoupled to avoid mutual interactions. Defects are randomly implanted within the bulk of

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