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Critical size and strength of the best bulk metallic glass former in the Mg–Cu–Gd ternary system

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We have developed a $Mg_{61}Cu_{28}Gd_{11}$ bulk metallic glass (BMG) that has the highest glass-forming ability among all the known ternary Mg-based BMGs. We also used a microcompression test to determine the intrinsic strength of such brittle Mg-based BMGs. The strength and plasticity of BMGs at small sample sizes are discussed in comparison with their behavior in conventional compression tests.

Keywords: Bulk metallic glasses; Magnesium; Microcompression; Strength; Plasticity

Amorphous alloys contain no well-defined dislocations that are responsible for yielding in crystalline materials, and are therefore expected to be strong and hard. The elevation of strength is particular desirable for the lightweight Mg alloys, which have relatively low strength compared with other engineering alloys. In recent years, bulk metallic glasses (BMGs) have been obtained in several Mg alloy systems [1–14]. However, as summarized in Table 1, all the monolithic Mg-based BMGs reported in literature are 'brittle', in the sense that their plastic strain to failure under compression is either nonexistent at all or very small (e.g., minor bending of the stress–strain curve, suggesting a plastic strain of a fraction of 1%). This causes two problems. First, due to the lack of plasticity, the apparent strength of these Mg-based BMGs is often irreproducible, one example of which is shown in Figure 1a for a $Mg_{61}Cu_{28}Gd_{11}$ BMG (see below for the details about this alloy). There is no obvious macroscopic yielding, and the failure occurs in the elastic region. The data scatter suggests that the failure stress is controlled by sample flaws [15], making it impossible to determine the intrinsic yield strength.

Secondly, the brittle behavior casts doubts on the usefulness of the Mg-based BMGs in structural applications, despite their high strength. In recent years, Mg-based BMG matrix composites have been developed where crystalline reinforcements have been used to impart observable plastic strains [15,16]. For the monolithic glasses, a recent discussion [17] suggests that the low ductility and the corresponding low process-zone size (see below) of BMGs would become unimportant when the sample dimensions are reduced down to the micrometer regime. This, together with the advantages of high strength/hardness and processing flexibility in the supercooled liquid region, may render (brittle) BMGs potentially usable for components in microelectromechanical systems (MEMS) [18–20].

The purpose of this letter is twofold: we report a new 12 mm BMG, the largest for all the ternary Mg-based BMGs, and show that this glass also has the highest strength of all the Mg-based BMGs published to date. The strength was determined using a compression test of microscale pillar samples. Sample size effects are expected for the strength and plasticity behavior of BMGs, on several different levels and for different reasons. First, in terms of strength, the probability of finding a flaw (such as pores, unmelted particles, oxides and inclusions) is low in a small volume of the BMG, such that one gets to measure the stresses needed to initiate plastic

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Table 1. Summary of previously published mechanical properties (fracture stress σ_f and plastic strain ε_p) of Mg-based BMGs, obtained in conventional compression tests

Alloy composition (at.%)	Cast sample diameter (mm)	σ_f (MPa)	ε_p (%)	Refs.
$Mg_{65}Cu_{20}Ag_{10}Y_2Gd_8$	1	956	0.3	[7]
$Mg_{65}Cu_{20}Ag_5Gd_{10}$	1	909	0.5	[9]
$Mg_{65}Cu_{15}Ag_{10}Gd_{10}$	1	935	0.2	[9]
$Mg_{65}Cu_{7.5}Ni_{7.5}Zn_5Ag_5Y_{10}$	1	832	0	[12]
$Mg_{65}Cu_{7.5}Ni_{7.5}Zn_5Ag_5Y_5Gd_5$	1	928	0.57	[12]
$Mg_{65}Cu_{15}Ag_5Pd_5Gd_{10}$	2	817	0.2	[8]
$Mg_{75}Cu_{15}Gd_{10}$	2	743	0	[10]
$Mg_{75}Cu_5Ni_{10}Gd_{10}$	2	874	0.2	[11]
$Mg_{65}Cu_{25}Gd_{10}$	2	834	0	[6]
$Mg_{65}Ni_5Cu_{20}Gd_{10}$	2	904	0.15	[6]
$Mg_{65}Cu_{7.5}Ni_{7.5}Zn_5Ag_5Y_{10}$	4	490–650	0	[15]
$Mg_{65}Cu_{15}Ag_5Pd_5Y_{10}$	5	770	0	[13]
$Mg_{61}Cu_{28}Gd_{11}$	4	461–732	0	This work
$Mg_{61}Cu_{28}Gd_{11}$	1	1075 ± 35	0.40	This work

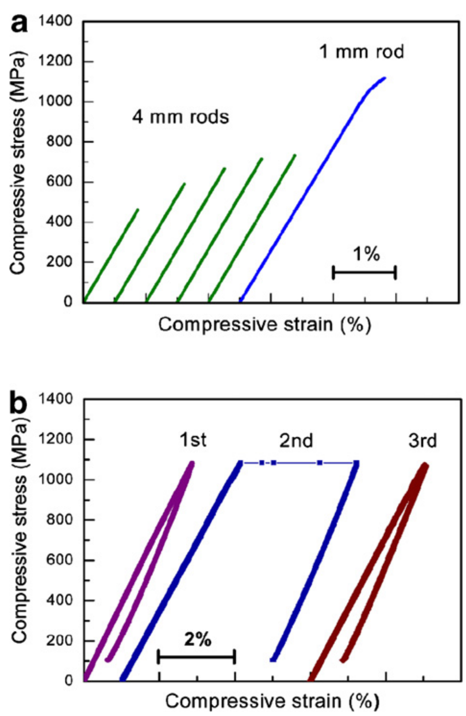

Figure 1. Engineering stress–strain curves obtained from (a) the uniaxial compression test of the 4 and 1 mm $Mg_{61}Cu_{28}Gd_{11}$ BMG, in comparison with that from the microcompression test in (b) microcompression stress–strain curves for the three loading/unloading cycles: the first loading cycle served as a trial test; the second loading revealed the plastic deformation; and the third loading confirmed the strength and plastic strain observed in the second loading.

flow (shear banding) [21,22], for the brittle BMGs in particular. As shown in Table 1, there has been no reliable measurement of strength for Mg-based BMGs. To observe any non-zero plasticity (usually very small, on the order of a fraction of 1% plastic strain; see Table 1), samples had to be cast directly into 1 mm diameter rods (barely a 'bulk' metallic glass). In these cases, of course, there may be effects due to the faster cooling rate and thermal history, which require separate studies. In other words, samples cut from Mg-BMGs, in the size range of typical compression tests (from one to several millimeters), have so far exhibited neither plastic strain nor consistent strength.

Second, in terms of plasticity, bending tests of metallic glass wires and thin plates usually indicate a considerable bending ductility [23]. If the onset of cracking is indeed suppressed [17] for a micro-sized deforming body, the shear offset achievable in even one single shear band may already result in large plastic strains. It would be interesting to observe the accommodation of the relatively significant (although nonuniform) plastic strains before failure when the BMG object is only micrometers in size. Third, for very small samples, the shear banding deformation mode may change altogether, causing major differences in strength and plasticity. This last point, however, requires submicron samples and separate investigations, and will not be dealt with in this short communication.

Our study uses a new BMG, $Mg_{61}Cu_{28}Gd_{11}$, which was discovered in a systematic search in the Mg–Cu–Gd ternary system. Among the $Mg_{65}Cu_{25}RE_{10}$ (RE = La, Ce, Pr, Nd, Sm, Gd, Dy, Tb, Ho, Er and Yb) ternary BMGs already studied, $Mg_{65}Cu_{25}Gd_{10}$ was reported to have the best glass-forming ability (GFA) [3]. A critical diameter (D_c, for copper mold casting) of 8 mm was achieved at this composition [2] by simply using Gd to substitute for Y in the $Mg_{65}Cu_{25}Y_{10}$ BMG ($D_c = 4$ mm [1]), which is known to have a eutectic composition. However, our experience has been that the GFA is strongly composition-dependent and the eutectic composition is not necessarily the best for GFA [4,5].

Elemental pieces with purity better than 99.9% were used as starting materials. Cu–Gd ingots as an intermediate alloy were prepared by arc melting under a Ti-gettered argon atmosphere in a water-cooled copper crucible. This alloy was then melted with Mg pieces by induction melting under inert atmosphere to obtain a master alloy with the nominal composition (in atomic percentage). The alloy ingots were melted several times to ensure compositional homogeneity. Then the master alloy was re-melted in a graphite tube using induction melting and injected in a purified inert atmosphere into the copper mold that has internal rod-shaped cavities about 50 mm in length. Chemical analysis of selected samples using inductively coupled plasma emission spectroscopy confirms the composition in the final alloy. The cross-sectional surfaces of the as-cast rods were analyzed by X-ray diffraction (XRD) using a Rigaku D/max 2400 diffractometer with monochromated Cu K_α radiation. Thermal analysis was carried out using a Perkin–Elmer differential scanning calorimeter (DSC-diamond) under flowing purified argon (graphite pans, with a heating rate of 20 K/min).

Compression test samples 8 mm in height were cut from the as-cast rods of 4 mm in diameter, prepared by suction casting. The loading surfaces were polished to be parallel to an accuracy of less than 10 μm. Uniaxial compression tests at room temperature were conducted using a constant strain rate of $1 \times 10^{-4}\ s^{-1}$.