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Tension behavior of metallic glass coating on Cu foil

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ABSTRACT

The tensile behavior of the monolithic ZrCu thin film metallic glass and the ZrCu/Cu multilayer coating on pure Cu foil is systematically examined. The extracted tensile modulus and strength of the 1 μm films are in good agreement with the theoretical rule of mixture prediction. The extracted 2 μm film data are lower, but can be corrected back by considering the actual intact cross-sectional area during the tensile loading. The current results reveal that the ZrCu/Cu multilayer coating exhibit much better tensile performance than the monolithic ZrCu coating despite of the brittle deformation. To obtain ductile amorphous/crystalline multilayer coating, two principles are suggested in this paper.

1. Introduction

Metallic glasses have attracted much attention over the past decade. Due to the amorphous atomic structure, metallic glasses exhibit unique mechanical and physical properties, such as high strength, high corrosion resistance, excellent shaping and forming ability in the viscous state and large elastic deformation about 2% and so on [1–4]. Among many metallic glass alloy systems, the Zr-based bulk metallic glasses (BMGs) possess outstanding mechanical properties [5–8]. Recently, thin film metallic glasses (TFMGs), prepared by sputtering, have received increasing attention as well [9]. In 1996, the synthesis of binary ZrCu TFMGs by using magnetron co-sputtering system in a relatively high vacuum was first investigated by Dudonis et al. [10]. In our group, the stabilized fabrication processes and structural identification of ZrCu TFMGs have also been examined [11]. It was found that the binary ZrCu TFMGs could be achieved over a much wider composition and substrate temperature range. Meanwhile, the simple composition and the rapid deposition process make the binary ZrCu system an ideal model for property analyzes and structural simulations [12].

The development of metallic glasses is still restricted by the crucial drawback of brittleness at room temperature, especially under tension [13]. ZrCu TFMGs, similar to their BMG samples, also show high strength and poor plasticity. Therefore, the approach to improve the ductility continues to be a significant issue. For BMGs, the improvements of plasticity in tension and compression have shifted to the concept of bulk metallic glass composites (BMGCs), by incorporating deformable metal phases into the metallic glassy matrix [14–17]. In parallel, the integration of amorphous and crystalline layers by adjusting their constituent has been regarded as a potential method for improving the ductility of metallic multilayer [18–20].

Wang et al. [18] demonstrated that a Cu/CuZr crystalline/amorphous nanolaminate with respective thickness of 35/5 nm reliably showed high strength and large tensile elongation before failure. The results indicated that the shear bands could be stopped by the crystalline layer and the dislocations could be disrupted by the amorphous layer. Furthermore, thick Cu nanocrystalline layers coupled with thin ZrCu TFMGs, deposited into a 100-μm-thick free-standing laminate composition, were studied by Nieh and Wadsworth [21]. This laminate exhibited a remarkable tensile elongation of 4%. Further investigations showed that the interplay of crystalline/amorphous layers not only suppress the shear band propagation and dislocation piling up but also provide the high defects capacity against the destruction [19,20]. Moreover, Liu et al. [22,23] systematically reported the results of microcompression tests on amorphous/nanocrystalline multilayer with 3 constituents of ZrCu/Cu, ZrCu/Mo and ZrCu/Zr with various layer thickness. The plastic compression strain can go up to more than 100% as a result of their multilayered structures.

In most previous studies, tensile testing on such amorphous–crystalline laminates was conducted on the free-standing specimens. However, it is difficult to perform tensile testing on multilayered films thinner than a few micrometers. In 1999, Macionczyk and Bruckner [24] reported the mechanical behavior of the 0.2–2.0 μm AlCu crystalline thin films deposited

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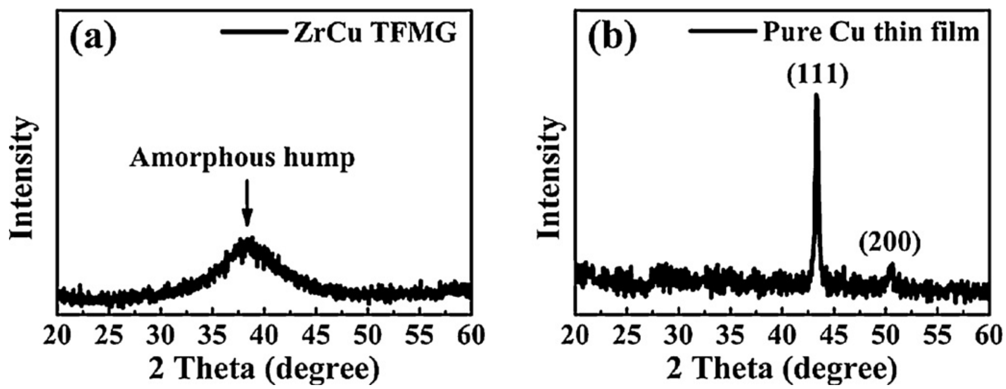

Fig. 1. The XRD patterns of: (a) the ZrCu TFMG deposited on the Si wafer and (b) the crystalline Cu thin film deposited on the Si wafer.

on the thin soft substrate such as the Kapton polyimide foil. The contribution from the polyimide foil could be extracted so that the intrinsic mechanical properties of thin film can be evaluated by simple calculations. Recently, Zhang et al. [25] also reported the tensile results of nanostructured soft-Cu/brittle-Cr multilayered thin films with equal individual layer thicknesses but different modulation periods, being coated onto the polyimide foils. For some tensile tests of thin films coated onto the polymer substrate, the large mismatch of the Poisson's ratios and thermal expansion between polyimide and TFMG might lead to complicated residual stress along the interface. In this study, in order to reduce the aforementioned interference, the monolithic ZrCu and nanolaminated ZrCu/Cu TFMGs are deposited on the pure Cu foils. These nanolaminates with the various layer and overall film thicknesses are systematically examined for their tensile characteristics.

2. Experiments

Amorphous/crystalline ZrCu/Cu multilayer thin films were deposited on the 10-μm-thick pure Cu foils, using a direct current (DC) magnetron sputtering system. For the tensile testing, the Cu foils were first cut into the dog-bone-shape by the laser patterning process with a pulsed UV laser before the sputtering processes. The dog-bone shaped specimens had a total length of 30 mm and a gauge section of 4.8 mm in length and 1.3 mm in width. In order to obtain a uniform layer thickness, the as-cut Cu foils were fixed on a rotated holder with a diameter of 50 mm at a speed of 15 rpm during the sputtering process. The working distance between the holder and the DC gun was set to be 120 mm. Then, a $Zr_{50}Cu_{50}$ (in atomic percent, at.%) alloy target and a pure Cu target were used for depositing the constituent layers of multilayer thin films. The base pressure of main chamber was pumped down to less than 5×10^{-7} Torr by a cryo-pump and pure argon atmosphere as the working gas was maintained at the rate of 30 standard cubic centimeters per minute (sccm). The DC powers of $Zr_{50}Cu_{50}$ and Cu guns were set to be 300 and 150 W, respectively.

The quantitative constituent analysis of each layer was conducted by an energy dispersive X-ray spectrometer (EDS) attached on the JEOL-6330 field-emission scanning electron microscope (SEM), to ensure the correct $Zr_{50}Cu_{50}$ and pure Cu compositions in each layer. To examine the nature of each layer, the as-deposited ZrCu and Cu layers on the Si wafer was also separately prepared and characterized by Siemens D5000 X-ray diffraction (XRD).

For the tensile testing of the Cu foils coated with the multilayer thin film, the ZrCu/Cu multilayer thin films with each layer thickness, h_i, of 100 nm but two total multilayered thickness, h_t, of 1 and 2 μm were deposited. This first depositing bottom layer onto the Cu foil is always crystalline Cu and the last top layer is ZrCu TFMG throughout. Uniaxial tensile testing with a force transducer of 250 N and a constant strain rate of $1 \times 10^{-3}\ s^{-1}$ was conducted on the coated specimens at room temperature by a MTS Tytron 250 Microforce Testing System (mini-tester). To detect the small displacements with high resolution, the variations in displacement were recorded by an axial displacement gauge assembly with the acquisition rate of 100 Hz. For comparison, the dog-bone-shaped Cu foils without coating were also tensile tested by more than 10 times under the same conditions to give reliable substrate data for subsequent subtraction bases. The Young's modulus and strength of the multilayer thin film were extracted by subtracting the contribution from the Cu foil in the coated specimen.

In addition, to analyze the layer thickness effects, tensile testing was also performed on the Cu foils deposited with 1-μm-thick ZrCu/Cu multilayer thin films with five different h_i values of 10, 25, 50, 100 and 250 nm. The failed specimens were machined by a SEIKO SMI3050 dual focus ion beam (FIB) system for observation of the cross-section and fracture surface morphology.

3. Results and discussion

To identify the structures of multilayer thin films, the XRD patterns of the ZrCu TFMG and crystalline Cu thin film deposited on the Si (1 0 0) wafers are shown in Fig. 1. Due to the amorphous nature, the as-deposited ZrCu TFMG presents a broad diffraction hump at the 2θ diffraction angle range of 30–45°, as shown in Fig. 1(a). Meanwhile, Fig. 1(b) exhibits the highly textured {1 1 1} planes of the as-deposited nanocrystalline Cu thin film, typical for most face centered cubic metallic films. Furthermore, the resultant grain sizes of as-deposited Cu layers with different thicknesses (10, 25, 50, 100 and 250 nm) were measured to be in the range from ten to several decades nanometer (30 ± 10 nm). For the 10-nm-thick Cu layer, the nanocrystalline grains are about 25 nm in diameter and 10 nm in height, exhibiting non-equiaxed shape. For the rest Cu layers with different thicknesses from 25 to 250 nm, the Cu grain sizes are all about 30 nm in diameter. It is postulated that the small variation of Cu grain size would impose minor influence on the tensile properties.

3.1. Theoretical prediction by rule of mixtures (ROM)

Before analyzing the experimental data, it is necessary to figure out the approximate right ranges for the tensile specimen modulus and maximum stress. The Young's moduli of the as-deposited crystalline Cu and monolithic ZrCu TFMG have been carefully measured by nanoindentation, as listed in Table 1. Based on these data, the theoretical Young's modulus E of ZrCu/Cu multilayer thin film