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Assessing the interfacial strength of an amorphous–crystalline interface

M.C. Liu [a], J.C. Huang [a,*], Y.T. Fong [b], S.P. Ju [b], X.H. Du [a,c], H.J. Pei [a], T.G. Nieh [a,d]

[a] Department of Materials and Optoelectronic Science, National Sun Yat-Sen University, Kaohsiung 804, Taiwan, ROC
[b] Department of Mechanical and Electro-Mechanical Engineering, National Sun Yat-Sen University, Kaohsiung 804, Taiwan, ROC
[c] School of Materials Sciences and Engineering, Shenyang Aerospace University, Shenyang 110034, PR China
[d] Department of Materials Science and Engineering, the University of Tennessee, Knoxville, TN 37996-2200, USA

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Abstract

In this study, an amorphous-ZrCu/crystalline-Zr nanolaminate (500 nm each layer) was initially synthesised using sputter deposition and then fabricated into micropillar samples using focused ion beam machining with the amorphous–crystalline (a–c) interfaces inclined 45° to the pillar axis. These pillars were subsequently tested in compression in order to study the response of a–c interfaces to the applied shear stress, and further compared with the one that tested with their a–c interfaces normal to the compressive direction. By combining the stress–strain behavior and electron microscopic observations of fracture mode, we were able to estimate the a–c interfacial strength. It was strong (~1.3 GPa), it could effectively transfer the load and it was capable of accommodating large shear strain, but it was not strong enough to suppress the interfacial sliding. Molecular statics simulations were also carried out to reveal the elastic–plastic behavior and, in particular, the deformation mode of the pillars. The computed results were in excellent agreement with the experimental observations.

Keywords: Metallic glasses; Compression test; Multilayer thin films; Fracture; Interface energy

1. Introduction

Structural and functional composites are widely used because of their balanced properties. Interfaces often determine the properties and thus the final performance of a composite material, especially when the characteristic dimension of the composite is small and the volume fraction of the interface region becomes dominant.

To understand the roles of the composite interface, the crystalline–crystalline (c–c) interface, whether it is metal–metal, metal–ceramic or ceramic–ceramic type, has been extensively examined [1–5]. In these cases, lattice mismatch, elastic strain and dislocation formation at the interfaces can be readily visualized and theoretically modeled. By contrast, amorphous–crystalline (a–c) interfaces are difficult to visualize and there has been very limited study on the structure and properties of such interfaces [6–14], yet many engineering applications are depending upon these interfaces for performance. For example, amorphous hard coatings on crystalline substrates are widely used for wear and fatigue resistance [15–17]. Recently, composite approaches were also adopted to improve the plasticity and toughness of intrinsically brittle metallic glasses (or amorphous alloys) [18–20]. In this case, a–c interfaces are pivotal on blocking and diverting shear band propagation to prevent catastrophic failure.

The lack of study on a–c interfaces probably results from the fact that structure and chemistry of such interfaces are largely uncertain. The analytical tools to investigate such interfaces are sophisticated and are generally required to have atomic resolution. Consistent preparation of test samples is also challenging. Recently, several groups [6,7,13] reported enhanced strength and plasticity in amorphous CuZr/crystalline Cu and Zr nanolaminates prepared by means of sputter deposition methods. These nanolaminates exhibit very high strength yet still retain a high tensile or compressive plasticity (>4%). The high ductility is

* Corresponding author. Tel.: +886 7 525 2000x4063.
E-mail address: jacobc@mail.nsysu.edu.tw (J.C. Huang).

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somewhat unexpected, since the amorphous layer is intrinsically brittle. In addition, the strength of the nanolaminate is significantly higher than that predicted by the rule-of-mixture. Both results indicate a strong interaction between the amorphous and crystalline layers via the interface. It is noted that these amorphous–crystalline nanolaminates were tested mainly in tension and compression. In tension, the samples were oriented with the interfaces parallel to the loading axis but, in compression, the interfaces were perpendicular to the loading axis. In either case, these interfaces were subject only to a normal stress without shear. The stress state at the interfaces obviously can affect dislocation operation or shear-band generation, and thus the strength and plasticity of a multilayer. In the present study, we prepared amorphous CuZr/crystalline Zr micropillar nanolaminates with the interfaces inclined 45° to the loading axis to investigate the mechanical response of the samples under pure shear.

2. Experimental and simulation methods

Multilayered thin films were synthesised by co-sputtering on Si substrate [8,10,13]. The base pressure prior to deposition was less than 5×10^{-7} torr and the work pressure was at 3×10^{-3} torr. The ZrCu layer is in a fully amorphous state, and the hexagonal close packed (hcp) Zr layer has the growth plane texture of basal (0 0 0 1). Both the ZrCu and Zr layers were fixed to 500 nm in thickness. The top layer was consistently the ZrCu TFMG film.

Microscaled pillar samples with inclined interface for compression tests were fabricated from the as-sputtered ZrCu/Zr nanolaminated films, using the Seiko SMI3050 dual FIB system equipped with the special handmade holder. The pillar measured ~0.9–1.0 μm in diameter and 2.4–2.5 μm in height, with a taper angle of ~2–3°. The JEOL scanning electron microscope (SEM) and transmission electron microscope (TEM) were selected to observe the microstructure and morphology of the pillar samples before and after microcompression testing. The micropillar samples were tested in uniaxial compression by using a commercially available nanoindentation system (MTS Nanoindenter XP). The flat-end Berkovich indenter machined by FIB has an equilateral triangle cross-section measuring 13.5 μm in side length.

To save computation time, we adopted molecular statics (MS) simulation, instead of molecular dynamics (MD) simulation, in this study. For MS simulations, all atomic interactions were modeled by the many-body tight-binding potential (TB) [21–24], and the form of potential was represented as

$$E_i = -\left\{\sum_j \xi^2 \exp\left[-2q\left(\frac{r_{ij}}{r_0}-1\right)\right]\right\}^{1/2} + \sum_j A \exp\left[-p\left(\frac{r_{ij}}{r_0}-1\right)\right]$$

where ξ is an effective hopping integral, r_{ij} is the distance between atoms i and j and r_0 is the first-neighbor distance. The parameters ξ, A, p, q and r_0 for Cu and Zr were determined on the basis of the experimentally obtained values of cohesive energy, lattice parameter, bulk modulus and shear elastic constants of Cu and Zr [25,26]. Table 1 lists parameters used for the potential. The material properties of Cu and Zr were accurately predicted by the TB potentials using these parameters [27,28]. To get an accurate modeling of Zr–Cu interaction for ZrCu metallic glass, the force matching method was used to determine the tight-binding potential parameters for the Zr–Cu interaction with the corresponding forces from the density functional theory (DFT) calculations [29]. Simulated results for the lattice volume, radius distribution and binding energies were very close to the experimental values, indicating that these DFT settings were accurate enough in predicting the material properties for amorphous ZrCu and crystalline Zr systems.

Before determining the mechanical properties of ZrCu metallic glass, the most important step is to obtain a reliable amorphous ZrCu configuration. In our basin-hopping (BH) method, the conjugate gradient method was replaced by the limited memory BFGS method (LBFGS, BFGS representing for four initiators, Broyden, Fletcher, Goldfaeb and Shanno) [30], which can be used to simulate a system consisting of a large number of atoms. Furthermore, the simulated annealing (SA) method was also implemented with the BH method to be a SABH method, which included a wider search within the energy space. The resulting simulated X-ray diffraction pattern was consistent with the experiment with a diffuse diffraction hump over $2\theta = 30$–45°.

MS simulation was subsequently employed to investigate the mechanical properties of ZrCu/Zr/ZrCu (top/middle/bottom) nanolaminates. Limited by simulation time, the simulated ZrCu/Zr/ZrCu nanolaminate pillar was much smaller than the experimental one, but still with the slight taper of 3°, top pillar diameter ~8.0 nm, bottom pillar diameter ~11.5 nm and pillar height ~35.0 nm. During the compression process, these structures are fixed by the substrate with a volume of $36 \times 36 \times 4\ \text{nm}^3$. The interface between ZrCu and Zr layers were modeled by three interfacial interaction energy conditions, namely the highly stable (HS), intermediately stable (IS) and less stable (LS) interfaces, which correspond to the well bonded (with low interfacial energy and high interfacial shear strength), intermediately bonded and poorly bonded (with high interfacial energy and low interfacial shear strength) interfaces, respectively. Although a MS study would provide the thermal properties corresponding to 0 K instead of the

Table 1
Parameters used in the tight-binding potential for MS simulation.

Interaction	A (eV)	Z (eV)	p	q	r_o (Å)
Cu–Cu	0.0783	1.2355	11.1832	2.3197	2.5560
Zr–Zr	0.1793	2.2014	9.3000	2.1000	3.1744
Cu–Zr	0.1935	1.2255	10.5158	2.0736	2.7067