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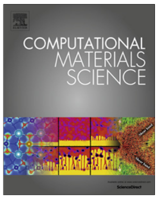

Plastic deformation mechanisms and size effect of $Cu_{50}Zr_{50}$/Cu amorphous/crystalline nanolaminate: A molecular dynamics study

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ABSTRACT

The plastic deformation behaviours of $Cu_{50}Zr_{50}$/Cu amorphous/crystalline nanolaminate were studied at the atomic scale using molecular dynamics simulations. Unlike the limited plasticity due to the highly localized shear banding in pure metallic glass, the nanolaminate undergoes a pronounced homogeneous deformation under applied tensile strains greater than 8.0% that is ~36.4% larger than that of pure metallic glass. This plasticity enhancement is attributed mainly to the suppression of the nucleation and propagation of the shear band in the nanolaminate. The scattered immature shear band or shear transformation zones and dislocations cooperate to form a network to transmit the strain within the entire sample. Significant size effects are demonstrated, revealing that both the strength and elongation-to-failure reach their respective peak values at a layer thickness d of approximately 4 nm and that the relationship between strength and d is well-explained by the confined layer slip (CLS) model. In the case of d values from 27 nm to 4 nm, the nanolaminate undergoes a transition from inhomogeneous deformation to homogeneous deformation. When d decreases to less than ~4 nm, the nanolaminate exhibits inhomogeneous deformation because of the destruction of the amorphous/crystalline interface. On the basis of this mechanical mechanism, the present study provides an approach for strengthening and toughening the amorphous/crystalline nanolaminate by adjusting the layer thickness.

1. Introduction

Over the past few decades, metallic glasses (MGs) have attracted considerable attention because of their high strength and hardness [1–4]. However, their near-zero tensile plasticity at room temperature has impeded the wide application of these materials [5–7]. Inspired by the laminar structure and the exceptional toughness of nacre (mother-of-pearl) [8], researchers designed the “architectured” material amorphous/crystalline nanolaminate (ACNL) by incorporating an MG and a crystalline material layer-by-layer at the nanoscale to improve the MG plasticity [9–15]. ACNL has been demonstrated to exhibit strong potential for combining high strength and good plasticity [12,13,16–19]. Nieh et al. [16] and Wang et al. [12] found that Cu_4Zr_3/Cu and Cu_3-Zr/Cu ACNLs have similar tensile strength values of approximately 1.1 GPa and that their elongation values reach 4% and 13.8 ± 1.7%, respectively. Kim et al. reported a fracture strain of 4% and a strength as high as 2.513 GPa for the $Cu_{50}Zr_{50}$/Cu ACNL [13].

The failure of pure MG is now well accepted as being induced by shear banding due to the accumulation of shear transformation zones (STZs) [20–24]; in addition, the plastic deformation of the pure crystal is known to be governed by dislocation or diffusionless solid-state phase transformations. However, in the architectured ACNL, the MG layer correlates with the crystalline layer via amorphous/crystalline interfaces (ACIs), and the plastic deformation behaviours of ACNL are thus much more complex than those of the free-standing pure MG and crystalline material. Compared to the experimental successes in the fabrication of ACNL, basic understanding of the microscopic details of ACNL plastic deformation lags far behind and is still open to study [12,13,25,26].

Although the *in situ* experimental observation of plastic deformation of ACNLs at the atomic scale is generally quite difficult, this level is most suitable for the use of molecular dynamics (MD) simulations [27,28]. Thus far, MD simulations have been extensively applied to characterize the shear band (SB) and dislocation behaviours of pure MG and crystalline materials [29–32]. However, only a few MD studies of the plastic deformation of ACNLs have been reported [12,33]. Wang et al. found that a dislocation could induce STZs when it impinges at the ACI [12]. Arman et al. demonstrated that the dislocation of the crystal layer could induce shear banding in the neighbouring MG layer [33]. In all of these simulation studies, the crystalline layer yields first during the plastic deformation of ACNLs. According to the Hall-Petch relation [34,35], the strength

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of a crystalline material will be greatly enhanced when its size is reduced to the nanoscale. Therefore, the possibility arises that the MG layer could yield before the crystal layer in an ACNL. Kim et al.'s observations of the $Cu_{50}Zr_{50}$/Cu ACNL provide some experimental evidence for this assumption. Currently, how SBs interact with dislocations in the ACIs if the MG layer yields first is unclear because Kim et al. did not discuss this deformation detail in their experimental work. However, the role of the ACI is still controversial and is an open question in the study of ACNL plastic deformation [25,26]. For example, Wang et al. proposed that the ACI acts as the source and sink of the dislocations and that they are therefore highly important [12]. However, Kim et al. concluded that ACI exerts no obvious effect on the plastic deformation [13]. Therefore, further works are needed to answer this question.

The effect of size on strength has been an important theme in nanolaminates research, and some relevant experiments have been reported in recent years [15,36,37]. In the traditional crystalline/crystalline nanolaminate (CCNL) structure, the Hall-Petch model is usually used to explain the size effect in the range from several hundreds of nanometres to 100 nm [36]. The Hall-Petch model applies to the deformation mechanism dominated by dislocations, where the dislocation pile-ups need to be continuous. However, when the thickness of the layer is reduced to tens of nanometres and the dislocations are more rare and no longer continuous, the results will deviate from the Hall-Petch relation. By contrast, the CLS model explains the strength size effect in the range from tens of nanometres to several nanometres, and involves the glide of single Orowan-type loops bounded by two interfaces [38–41]. When the CCNL size decreases to several nanometres, dislocations penetrate and transmit across the interfaces, resulting in the destruction of the interface. The size effect for ACNL differs from that for the CCNL structure because of the absence of the normal crystal/crystal coherence or semi-coherence relation at the interface between the crystal and the MG. Guo et al. [15] and Zhang et al. [37] used the CLS model to explain and fit the size effect of the ACNL quite well. Nevertheless, more detailed information and theoretical studies are necessary because of the particularity and complexity of ACNL.

The results obtained by the simulations described in this paper provide a useful approach for the preparation of a strong and ductile ACNL by designing an optimal structure with a proper size and aspect ratio of individual layers. Research emphasis is placed on the microscopic details of the plastic deformation process of an ACNL and the size effect of the mechanical properties. Our analysis demonstrates that laminate structures and ACIs play substantial roles in enhancing the tensile ductility.

2. Simulation method

The MD simulations were performed for $Cu_{50}Zr_{50}$/Cu ACNLs with a time step of 2 fs. The interatomic interactions are described by the embedded-atom-method (EAM) potential proposed by Sheng et al. [42,43]. All simulations were completed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [44] with "ovito" as a visualization and auxiliary analysis tool [45].

To prepare the nanolaminate, we first fabricated a large MG sample of 3,500,000 atoms by vitrifying a $Cu_{50}Zr_{50}$ melt equilibrated at 2000 K with a cooling rate of 10^{12} K/s in the isothermal and isobaric (NPT) ensemble. The temperature and pressure were controlled according to the Nose-Hoover method [46,47]. The glass-transition temperature was determined as 705 K from the temperature dependence of enthalpy. After aging at 50 K for 0.5 ns, an MG film was cut from the large MG sample following the standard of minimizing the mismatching stress and was then merged with crystalline Cu, which was also well equilibrated at 50 K. The [1 0 0] direction of the Cu crystal was perpendicular to the ACI. To release the stress, the whole nanolaminate was heated to 600 K at 10^{12} K/s, aged at 600 K for 0.5 ns, and then cooled to 50 K at 10^{12} K/s.

To elucidate the effects of the size and aspect ratio, we prepared nine samples, including one pure MG sample and eight ACNL samples named for convenience as w_1/w_2, where w_1 and w_2 denote the thicknesses of the MG and crystalline layers, respectively. In this work, we let $w_1 = w_2 = d$, where d was 27.1, 13.4, 9.0, 6.8, 5.5, 4.0, 3.2, and 1.8 nm. The atomic models for the pure MG and the ACNLs are illustrated in Fig. 1. The pure MG sample contains ~900,000 atoms, and the ACNL samples are composed of ~1,100,000 atoms; the samples show similar sizes of $54 \times 5.4 \times 54\ \text{nm}^3$. During the deformation process, periodic boundary conditions (PBCs) were used along the x- and y-axes, and the free surface condition was applied for the z-axis. A constant strain with a rate of $4.0 \times 10^7\ \text{s}^{-1}$ along the x-direction was imposed. The stress was calculated by the component of the *virial* stress along the x-direction.

3. Results and discussion

3.1. Size effect of mechanical properties under tensile loading

The tensile stress-applied strain curves of the ACNLs and pure MG are illustrated in Fig. 2. On the basis of the standard linear response theory [48], the stress tensor is calculated as

$$\sigma_{\sigma\beta} = -\frac{1}{V}\sum_i \left[\frac{P_{i\alpha}P_{i\beta}}{m_i} + \sum_{j<i} r_{ij\alpha} f_{ij\beta}\right], \quad \alpha, \beta \in [1,3] \tag{1}$$

where V is the system volume, $P_{i\alpha}$ is the α-component of the momentum for atom i with mass m_i, and $f_{ij\beta}$ is the β-component of the force between atoms i and j separated by distance r_{ij}. As shown in Fig. 2, the ACNLs with different architectured structures show elastic modulus values similar to that of pure MG. This means that the architectured structure of ACNL has little effect on the elastic properties. However, for the same volume fraction of the crystalline phase, the architectured structure strongly influences the ACNL strength and tensile plasticity. On the one hand, the maximum strengths of all ACNL samples studied in our simulations are greater than that of pure MG. The ACNL 4 nm/4 nm is the strongest one, with a strength-to-failure, σ_f, of 3.13 GPa, which is approximately 34% higher than the 2.34 GPa value of pure MG. On the other hand, a proper ACNL architecture could greatly improve the tensile plasticity. For example, the elongation-to-failure, ε_f, of ACNL 4 nm/4 nm is ~8.02%, surpassing the 5.88% value of pure MG by approximately 36.4%. Therefore, the tensile strength and the tensile plasticity of MG are improved simultaneously in this work by the fine-tuning of the ACNL structure. This improvement is quite meaningful for the strengthening and toughening of materials because, in the normal case, an increase of the strength is often accompanied by a degradation of the plasticity. ACNL 27.1 nm/27.1 nm, in particular, is special in that its elongation-to-failure is apparently lower than those of other ACNLs and is even lower than the value for the pure MG; this phenomenon will be explained in the later sections.

The changes in the σ_f and ε_f of the ACNLs with d are presented in Fig. 3. The simulations were repeated five times for each sample. The maximum relative deviations of the strength of failure and the elongation to failure are 1.13% and 2.20%, respectively. Obviously, both of these mechanical properties exhibit a strong size effect. With decreasing d, the σ_f of the ACNL increases rather rapidly and reaches a peak value of 3.13 GPa at $d = 4$ nm. A further decrease of d leads to a decrease of σ_f. Similarly, the increase of ε_f stops when d is reduced to 4 nm. Therefore, the ACNL softens when