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Scale-dependent fracture mode in Cu-Ni laminate composites

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Abstract

Fracture behavior of Cu-Ni laminate composites was investigated by tensile testing. It was found that as the individual layer thickness decreases from 100 to 20 nm, the resultant fracture angle of the Cu-Ni laminate changes from 72° to 50°. Cross-sectional observations reveal that the fracture of the Ni layers transforms from opening to shear mode while that of the Cu layers keeps shear mode. Competition mechanisms were proposed to understand the variation in fracture mode of the metallic laminate composites associated with length scale.

Keywords: Fracture; nanolaminate; length scale; interface

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1. Introduction

A material with laminate structures often has enhanced mechanical properties compared with the constituent materials. For example, the hardness or yield strength of Cu-X (X = Ag, Cr, Ni, Nb, etc.) nanolaminates is significantly higher than that of the Cu film of the same thickness [1-3]. The improved mechanical properties of the laminate composites are related to the layer confinement on dislocation activity, the modulus mismatch, the lattice mismatch, and the coherent stress in the layers. Especially, interfaces in laminate materials may become dominant factors to control deformation behavior of the laminate structure as their geometrical and/or microstructural scale reduces to the nanometer regime [4]. It is expected that the optimum mechanical properties of the laminate structure with both high strength and good ductility may be obtained by the reasonable selection of the constituents with different mechanical properties, the interface between the constituents as well as the laminate scale [1-5]. Even though a number of investigations have focused on the yield strength of the nanolaminates based on the concept of dislocation mobility in the confined geometry, the microscopic process of fracture and the physical mechanism of the material associated with length scale and interface are still less understood. In this paper, we report the examination of fracture behavior of Cu-Ni laminate composites with individual layer thickness ranging from 20 to 100 nm. Layer thickness dependent fracture mode was found and the physical mechanism was analyzed.
2. Experimental procedure

Cu-Ni laminates with a nominal total thickness of 750 nm were deposited onto a dog-bone-shaped polyimide substrate by radio-frequency (RF) magnetron sputtering system. The thickness of the flexible substrate is 125 μm. The individual layer thickness (λ) of the Cu layer are selected to be 20, 50 and 100 nm while that of the Ni layer is 1/2 of the thickness of Cu layer. To ensure that the three types of Cu-Ni laminates have the same total thickness, the number of bilayers deposited is 25, 10 and 5, respectively. Before the deposition, the substrate was firstly cleaned by Argon plasma at a RF power of 50 W and a pressure of 0.4 Pa for 5 minutes. Then, a Ni layer was firstly deposited onto the polyimide substrate and the final top layer of the multilayer was ended by the Cu layer.

Tensile tests of the dog-bone-shaped sample of the Cu-Ni laminate/polyimide composites with a gauge section of 15 mm in length and 5 mm in width were performed by using a micro-force testing machine. All the samples were stretched to a total strain 5% at a constant strain rate of $1.6 \times 10^{-4}$ s$^{-1}$ and then unloaded. The planar and the cross-sectional views of the Cu-Ni laminates were examined by a dual-beam focused ion beam/scanning electron microscope (FIB/SEM).

3. Results and discussion

The surface morphologies of Cu-Ni laminates after tensile test were examined by an SEM. Multiple cracks formed on the surface of all specimens with a regular spacing. A typical surface morphology of the $\lambda=100$ nm Cu-Ni laminate after tensile
testing is shown in Figure 1a. It is found that channel cracks were formed and propagated along a direction nearly perpendicular to the loading direction. The median crack spacing is about 30 μm. Figure 1b reveals that the 50 nm Cu-Ni laminate was fractured in a shear mode.

To evaluate the detailed fracture behavior of Cu-Ni laminates in the through-thickness direction, FIB cross-section milling was conducted to have images of the cracking morphology. Figures 2a-2c present the SEM cross-sectional images of the cracks in the \( \lambda = 100, 50 \) and 20 nm laminates, respectively. A close inspection of the cracking morphology of the \( \lambda = 100 \) nm Cu-Ni laminate (Figure 2a) reveals that fracture modes in the Cu and Ni layers are shear and opening, respectively. In contrast, both Cu and Ni layers in the \( \lambda = 20 \) nm Cu-Ni laminate experience shear mode fracture. A high magnification image of the squared region in Figure 2c is shown in Figure 2d. A shear offset \( (\delta) \) of about 30 nm along the fracture plane has occurred at the interface between the Cu-Ni laminate and the polyimide substrate. The value of \( \delta \) varies with the distance from the cross-section position to the crack tip. Similar phenomenon was observed in the deformation zone ahead of the crack tip in the Cu-Ta multilayer [6].

A resultant fracture angle \( (\theta) \) is defined as the angle between the macro-cracking direction and the horizontal direction shown in Figure 2a. The values of \( \theta \) are measured statistically based on the cross-section images as that shown in Figures 2a-2c and present in Figure 3 as a function of \( \lambda \). It is evident that the value of \( \theta \) decreases from 72° to 50° with decreasing \( \lambda \) from 100 to 20 nm.
In light of the above observations, the macroscopic deformation process of Cu-Ni laminates may be understood as follows. In the initial stage of loading, both Cu and Ni layers of the Cu-Ni laminate undergo elastic deformation. With increasing the applied strain, the softer Cu layer yields first and the dislocations in the Cu layer will be activated and pile up at the Cu-Ni interfaces. Owing to differences in elastic modulus and strength between the Ni and Cu layers, the local stress in the Ni layer will increase much faster than that in the Cu layer as the applied strain further increasing. This would lead to the stress incompatibility at the interface. A continuous increase of the applied strain during tensile loading would finally cause premature fracture of the Ni layer while the Cu layer at the moment is still subjected to plastic deformation.

Microscopically, the observed variation of fracture mode (Figure 2) may be associated with length-scale dependent dislocation activity in the confined layers. When the Cu layer thickness is larger, the number of the dislocations piling up at the interface becomes much more so that the concentrated stress created by the dislocation pile-up is large enough to cause the harder Ni layer fracture. With the decrease in the layer thickness the number of dislocations in the pile up becomes fewer and fewer due to the limited gliding distance and the strong confinement of layer thickness on dislocation motion. When the layer thickness is smaller than a certain value, weaker or no dislocation pile-up will be created at the interface. As a result, the plastic deformation of the Cu-Ni laminate mainly involves glide of single Orowan-type dislocation loops in the Cu layer. It is expected that at this length scale
regime, the fracture behavior is dominated by the mechanism of the dislocation
crossing interface [Misra]. In what follows, we will estimate the upper and lower
limits of layer thickness, which determines the variation of fracture behavior of Cu-Ni
laminates.

We first estimate the upper limit of the layer thickness where dislocation pile-up

\[ \sigma_p = \frac{2\pi\sigma^2(1-\nu^2)h'}{Eb} \]  

where \( \sigma \approx 1 \text{ GPa} \) is the applied stress, \( E(=110 \text{ GPa}) \), \( \nu(=0.324) \) and \( b(=0.256) \) are
Young’s modulus, Poisson’s ratio and Burgers vector of Cu, respectively, and
\( h' = \lambda/\sin\phi \) is the length of the pileup, \( \phi(=70.5^\circ) \) for a <111> out-of-plane oriented grain
is the angle of between the slip plane and the interface. On the other hand, the
dislocation pile-up at the Cu-Ni layer interface can also be regarded as a crack with a
length equal to \( h' \) in the Cu layer. The fracture strength (\( \sigma_f \)) of the “cracked” Cu-Ni
laminate can be calculated from the Griffith fracture criterion [11] as

\[ \sigma_p = \sqrt{\frac{2\pi E\gamma}{h'}} \]  

where \( E(=208 \text{ GPa}) \) and \( \gamma(\approx 2 \text{ J/m}^2) \) are the Young’s modulus and the surface energy
of Ni, respectively. It should be noted that the Griffith fracture criterion is only valid
for brittle fracture (or small scale yielding). In the present case, the measurement of
the thickness of Ni layer before and after tensile test from the SEM cross-section images (Figure 2a) indicates that there is no significant change in the Ni layer thickness. This result means that plastic deformation in the Ni layer can be neglected upon using the Griffith criterion.

By using the above parameters and Equations (1) and (2), $\sigma_P$ and $\sigma_F$ is shown in Figure 4a through plotting $\sigma_P$ and $\sigma_F$ normalized by Young’s modulus as a function of $\lambda$. A transition point of $\lambda_U \approx 92$ nm was found at an intersection point of the two curves. This indicates that as $\lambda > \lambda_U$ corresponding to region I in Figure 4a, $\sigma_P$ will be larger than $\sigma_F$ of the Ni layer. That would result in mode I (opening) fracture of the Ni layer. The combination of the shear fracture of the Cu layer along the slip plane and the opening fracture of the Ni layer resulted in a macro crack, as depicted in the lower-right inset in Figure 3. As the Cu-Ni laminate has a (111) texture[], the slip system with the largest Schmid factor has a tilt angle of 70.5° to the interface. Taking the thickness ratio of the Cu to the Ni layer (about 2:1) and the different fracture modes of the Cu and Ni layers into consideration, the measured fracture angle (72°) based on Figure 2a for the $\lambda=100$ nm Cu-Ni laminate is consistent with the tilt angle of 70.5° of the slip plane. Thus, it is believed that the fracture of the Cu-Ni laminate with the larger $\lambda$ (region I in Figure 4a) is controlled by the dislocation pileup mechanism.

As $\lambda$ decreasing, the stress needed to drive dislocations motion in the layer channel is increased due to the strong constraint of the nanoscale layer thickness. This can be supported by the previous observations of the increase in yield strength with
decreasing $\lambda$ of the Cu-Ni laminate [13]. As a result, there are not enough dislocations to be activated in the same slip plane of the Cu layer. Only few dislocations or single dislocation could not establish a strong pile-up to generate enough stress concentration at the Cu-Ni layer interface to cause the opening mode fracture of the Ni layer. However, when the applied shear stress to drive dislocation motion becomes larger than the barrier strength of the layer interface, the interface is expected to be directly sheared and led to a shear offset, as observed in Figure 2d and depicted in the upper-left inset in Figure 3.

The lower limit of the layer thickness for the onset of the dislocation crossing interface can be evaluated as follows. Given that dislocations in the layer channel would not transmit across the interface until the stress for a dislocation motion is larger than the layer interface barrier strength, the interface barrier strength ($\tau_{bs}$) can be evaluated by considering both lattice mismatch and shear modulus mismatch of a metallic multilayer system as follows [6],

$$\tau_{bs} = \tau_\eta + \tau_G$$

The first term ($\tau_\eta$) in the right hand of Equation (3) represents the maximum barrier strength of an interface to the glissile dislocation movement due to the lattice mismatch [14], while the second term ($\tau_G$) reveals the barrier strength of an interface due to the shear modulus mismatch between two constituents [15]. Furthermore, the applied shear stress ($\tau_{ap}$) needing to have a dislocation loop glide in the confined layer is estimated to be inversely proportional to $\lambda$ [16, 17]. The values of $\tau_{bs}$ and $\tau_{ap}$ as a
function of $\lambda$ are plotted in Figure 4b. For the present Cu-Ni laminate, the intersection point of the $\tau_{ap}$ curve with the $\tau_{bs}$ line yields a critical individual layer thickness $\lambda_L \approx 21$ nm [6], which is much smaller than $\lambda_U$. As a result, two significant fracture mechanisms may be figured out at different length scales. For $\lambda > \lambda_L$, especially $\lambda > \lambda_U$, the fracture of the layer is dominated by the stress incompatibility between two constituent layers and dislocation pileup-induced stress concentration at the layer interface. While for $\lambda < \lambda_L$ corresponding to the region II in Figure 4b, the dislocations would transmit across the interface and result in a shear offset, which eventually causes the shear mode fracture of the Ni layer. The predicted $\lambda_L$ is quite close to the individual layer thickness of Cu-Ni laminate composites where shear mode fracture occurred in both the Cu and the Ni layers. This result indicates that the decrease in the length scale of the laminate structure leads to the prevalence of the shear-mode fracture, and that may directly have an influence on deformation stability and plasticity of the laminate composites.

4. Conclusions

In summary, we found that the resultant fracture angle of the Cu-Ni laminate gradually decreased from 72° of the $\lambda=100$ nm laminate to 50° of the $\lambda=20$ nm laminate. The fracture behavior of the Ni layer in the Cu-Ni laminate transforms from opening to shear mode as the Ni layer thickness is decreased from 50 to 10 nm, while the Cu layer always shows a shear mode fracture. A competition behavior between dislocation mobility inside the confined layer and the interface strength of the
constituent layer plays an important role in the change of fracture mode with $\lambda$.

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References


Figure Captions

Figure 1. SEM images of the surface of the Cu-Ni laminate after tensile test. (a) Parallel channel cracks in the $\lambda=100$ nm Cu-Ni laminate. (b) The inclined image (of tilted angle 52°) indicating shear-mode fracture in the $\lambda=50$ nm Cu-Ni laminate.

Figure 2. Cross-section SEM images of the fractured Cu-Ni laminate. (a) $\lambda=100$ nm Cu-Ni laminate, showing opening-mode fracture in the Ni layer and shear-mode fracture in the Cu layer. $\theta$ is defined as the resultant fracture angle. (b) and (c) The fracture images of Cu-Ni laminates with $\lambda=50$ and 20 nm, respectively. (d) A high magnification image of the area framed by the rectangle in (c) showing a shear offset ($\delta$) of about 30 nm as indicated in the image.

Figure 3. The variation of fracture angle of Cu-Ni laminates as a function of $\lambda$. The bottom-right inset schematically shows that in the thicker layer regime dislocation pileup at the Cu-Ni interface results in opening-mode fracture of the Ni layer. The upper-left inset illustrates shear-mode fracture occurring both in Cu and Ni layers.

Figure 4. (a) The dislocation pileup-induced concentration stress ($\sigma_p$) and the fracture strength ($\sigma_F$) normalized by Young’s modulus as a function of $\lambda$. (b) The interface barrier stress ($\tau_{bs}$) and the applied stress ($\tau_{ap}$) for a dislocation loop glide in the confined layer as a function of $\lambda$. The region I and region II in Figure 4 indicate the regime where opening-mode fracture and shear-mode fracture is prone to occur, respectively.
Figure 1

Figure 2
Figure 3

Figure 4