

Bilayer thickness effects on nanoindentation behavior of Ag/Ni multilayers

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The effects of bilayer thickness on the mechanical behavior of Ag/Ni nanomultilayers have been investigated by nanoindentation hardness and creep tests. The hardness increased with decreasing bilayer thickness, although as the bilayer thickness decreased below 8 nm there was a decrease in hardness that correlated well with increasing creep rate. The dependence of the creep rate on the bilayer thickness reveals that the grain boundary deformation is more dominant at bilayer thicknesses below 8 nm.

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Multilayers with periodicity in the nanoscale range have attracted much attention due to their enhanced mechanical properties compared to their bulk counterparts [1]. This enhancement has been explained by the Hall–Petch model, which is based on dislocation pile-up [1–4]. Recently, several studies have reported a breakdown in Hall–Petch behavior when the bilayer thickness, λ , is of the order of a few nanometers, but these softening mechanisms have not been completely clarified [2,3]. Molecular dynamics (MD) simulations with nanocrystalline materials have shown that the unusual mechanical behavior below the critical size is attributed to grain boundary sliding and Coble creep [5–7]. Despite the crucial roles of grain boundaries in the nanoscale range, few experimental studies have examined the time-dependent diffusional process in nanoscale multilayers. From this perspective, it is necessary to characterize the creep response of nanoscale multilayers at high strain rates and ambient temperature. To elucidate the underlying deformation mechanism in nanoscale multilayers, we present here the results of nanoindentation creep tests on Ag/Ni nanomultilayers with various bilayer thicknesses.

Multilayered Ag/Ni films with Ag and Ni layers of equal thickness were prepared by DC magnetron sputtering at room temperature under an argon pressure of 3.0 mTorr. Seven samples with different bilayer thickness

were made: 3, 5, 8, 10, 20, 50 and 100 nm. These samples were alternatively deposited on p-type Si (100) at deposition rates of 0.47 nm s^{-1} for Ag and 0.40 nm s^{-1} for Ni. The total thickness was usually of the order of $5 \mu\text{m}$. Cross-sectional transmission electron microscopy (TEM) micrographs of the samples showed a perfect layered structure with sharp planar interfaces between the two phases as shown in Figure 1a. Each layer consists of a polycrystalline columnar structure in which the in-plane grain dimension is of the order of the layer thickness, indicating that the grain size, d , is one-half of the bilayer thickness:

$$d = \lambda/2. \quad (1)$$

Using the grazing incidence X-ray diffraction (GIXRD) technique, we confirmed that the measured periodicities matched the designed bilayer thickness well, as shown in Figure 1b.

The multilayer hardness was measured using a nanoindenter (Triboscope; Hysitron, Minneapolis, MN). A series of indents was made on each sample using maximum loads between 100 and $3000 \mu\text{N}$. The maximum load was kept below a contact depth of 500 nm to avoid the substrate effect, which is negligible at below one-tenth of the total film thickness. A nanoindentation creep test was used to study the creep behavior of nanoscale multilayers. A $20 \mu\text{m}$ radius 90° conical indenter tip was used to apply uniform stress. The load was increased to maximum loads between 1000 and $8000 \mu\text{N}$ for 3 s, held for 60 s, and then unloaded for 3 s.

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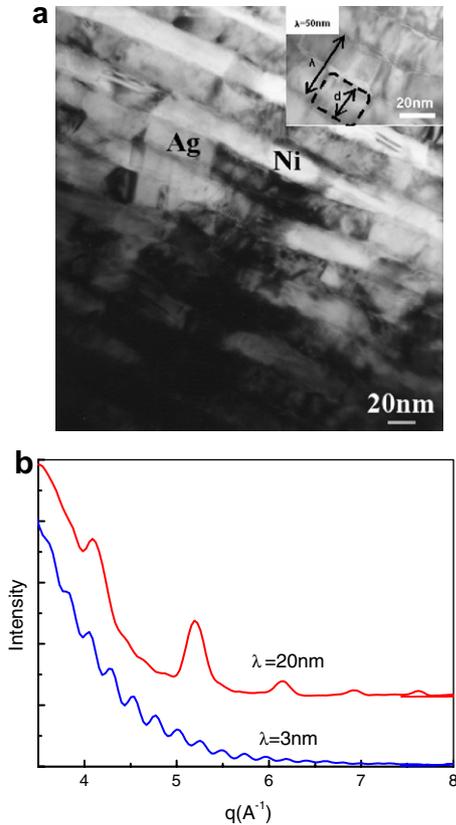


Figure 1. (a) Typical cross-sectional TEM micrograph ($\lambda = 50$ nm) and (b) GIXRD spectra of Ag/Ni nanomultilayers with various bilayer thicknesses. The inset is the enlarged TEM micrograph showing that every grain is confined to the layer thickness. The GIXRD spectra are displayed for scattering vector, q .

Figure 2 shows the hardness of the multilayers as a function of the bilayer thickness. The hardness increased as the bilayer thickness decreased (region I), although below a bilayer thickness of 8 nm, the hardness decreased as the bilayer thickness decreased (region II). Similar trends have been observed in previous works on various multilayers systems [1–3]. Typically, the bilayer thickness is substituted for grain size when characterizing the mechanical properties of multilayers. The hardness enhancement shows a good agreement with the Hall–Petch relationship (dashed lines in Fig. 2)

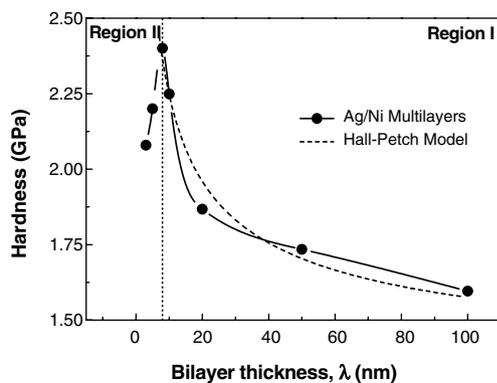


Figure 2. Hardness of Ag/Ni multilayers as a function of bilayer thickness. The dashed curve shows the Hall–Petch relationship.

which indicates that dislocation pile-up is the major strengthening mechanism. However, the breakdown of the Hall–Petch behavior at thicknesses below 8 nm implies that a typical pile-up-based dislocation model no longer controls the deformation. MD simulations indicate that the interface barrier to slip transmission decreases as the dislocation core dimension approaches the layer thickness [8]. Interface crossing at a characteristic interface barrier strength independent of the layer thickness is a possible operative mechanism. In addition, the effects of other factors such as elastic inhomogeneity, residual stress and image stress are thought to explain the multilayer strength at all length scales [9].

Figure 3 shows the change in the indentation depth during the holding period as a function of time in the nanoindentation creep test. The indentation depth increases with time and the rate of change with depth increases significantly with time below 10 nm. Ag/Ni multilayers with a small bilayer thickness showed a high creep response even at room temperature, indicating that the mechanism underlying the softening regime is closely related to time-dependent deformation. Based on the relationship between indenter geometry and the related mechanical behavior, the indentation strain rate has typically been defined as instantaneous displacement rate/instantaneous displacement or \dot{h}/h [10,11]. The strain rates were calculated by taking the time derivative of the displacement during the hold period under a constant load and dividing it by the displacement at that point. As the hardness $H(=P/A)$ represents the mean stress during indentation, the average hardness during the hold period has been used for stress in creep tests at a constant rate of loading/hold indentation [10].

The increase in the nanoindentation creep rate as a function of the applied stress at different bilayer thicknesses is shown in Figure 4. To avoid surface instability and substrate effects in the nanoindentation test, the range of applied stress for the proper indentation depth should be limited and differ in each sample. This tendency for the strain rate to increase with decreasing bilayer thickness is very similar to the increase in the creep rate seen with temperature in nanocrystalline Cu [12]. When diffusional creep is the significant mechanism in nanocrystalline materials with a high grain boundary volume fraction, the decrease in bilayer thickness has a

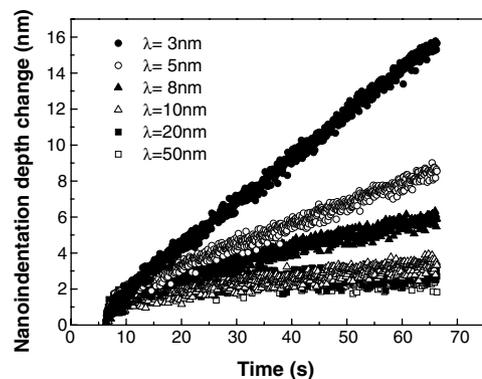


Figure 3. Creep response of Ag/Ni multilayers with different bilayer thicknesses at room temperature. The depth changed from the initial depth over the 60 s hold period under a constant load of 5000 μ N.

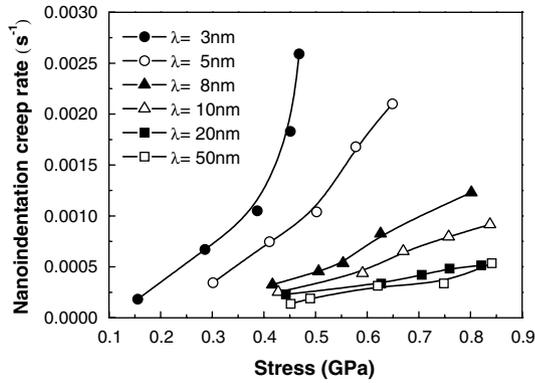


Figure 4. Nanoindentation creep rates as a function of the applied stress with different bilayer thicknesses at room temperature.

similar effect to the temperature increase because the creep rate is increased by a diffusion-activated process. Our nanoindentation creep results performed at constant room temperature show that the grain boundary-related parameter mainly affects the variation in the creep rate. Consistent with other creep results in nanocrystalline materials [12–14], the dependence of stress on the creep rate was observed for the entire range of bilayer thicknesses investigated.

The creep rate data at 0.47 GPa (an available stress value common to all samples) extracted from Figure 4 are shown in Figure 5 as a function of bilayer thickness. The nanoindentation creep rate increased significantly in the region where the hardness decreased. Above a critical bilayer thickness of 8 nm, the strain rate becomes less dependent on the bilayer thickness, indicating a transition into a different regime. Evidence of an obvious crossover in the deformation mechanism at a bilayer thickness of 8 nm is seen in the different magnitudes of the slope in the plot of $\log \dot{\epsilon}$ vs. $\log \lambda$. The data points for the three smallest bilayer thicknesses fall on a straight line with a slope of 1.938, i.e. λ^{-2} dependence on the indentation creep rate. It is well established that individual creep mechanisms that exhibit strain rates dependent on stress, temperature and grain size can be described by the Mukherjee–Bird–Dorn (MBD) constitutive equation [15]:

$$\dot{\epsilon} \propto d^{-p} \sigma^n \quad (2)$$

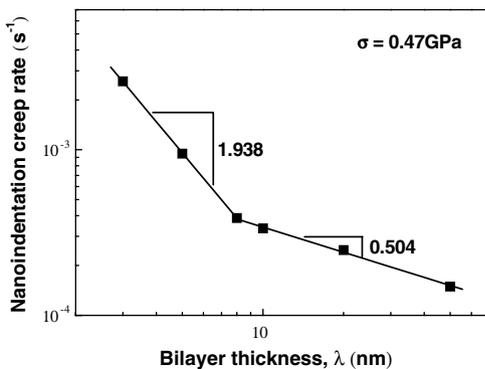


Figure 5. Creep rates of Ag/Ni multilayers as a function of bilayer thickness. The data were extracted from Fig. 4 at $\sigma = 0.47$ GPa.

Based on the general relationship, the stress exponent value, n , and the grain size exponent, p , can be used to identify the mechanisms controlling the deformation process. Several uncertainties in the nanoindentation creep tests have arisen because much of the material under stress is likely to be in the primary creep regime and the high gradients of stress and strain rate near the indenter. Although the nanoindentation creep parameter does not equal the typical steady-state creep parameter, the λ^{-2} dependence (substituting λ for d) of the stress exponent on the indentation creep rate below a bilayer thickness of 8 nm is related to the transition of the grain boundary deformation mode. The sudden increase in the stress exponent indicates that the grain boundary deformation governs the creep behavior below the critical thickness, whereas the deformation behavior at larger thicknesses is caused by a dislocation process, as shown in Figure 2.

The softening seen in most nanomultilayer systems has been in the bilayer thickness range of 2–10 nm while the softening of nanocrystalline materials has been observed at grain sizes of 10–50 nm [3,16]. The critical bilayer thickness is determined by the intersection of the flow stress given by the constitutive equation for interface hardening with that for interface softening. Irrespective of interface cutting mechanism [9] and thermally activated grain boundary shear mechanism [16], the grain boundary provides the main route for softening in the thickness range of a few nanometers. Therefore, a lower limit to critical bilayer thickness is determined when either the spacing between dislocations due to elastic interactions becomes larger than the layer thickness or the flow stress reaches the theoretical strength. Previous MD simulation results show that significant grain boundary motion is associated with large levels of deformation strain in nanocrystalline metals, although the grain boundary migration occurring in conjunction with dislocation emission is a necessary condition for grain boundary motion [8]. Furthermore, the higher fraction of grain boundaries provides a high diffusivity path [17] and the Frank–Read source is inoperative below the critical bilayer thickness [18]. As a result, the high creep rate and the decrease in hardness at very small bilayer thickness is due to the decreased flow stresses associated with grain boundary deformation, as shown in Figure 6.

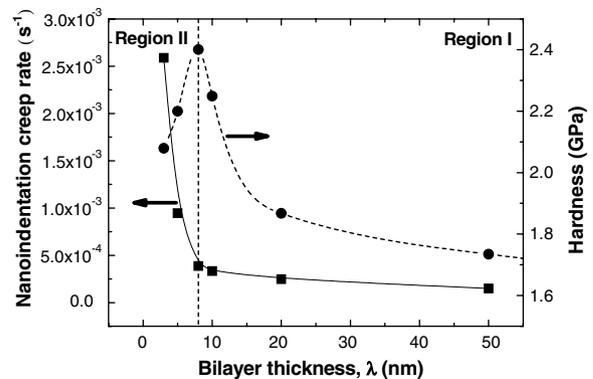


Figure 6. Comparison of the nanoindentation hardness and creep rate (at $\sigma = 0.47$ GPa) of Ag/Ni multilayers as a function of bilayer thickness.

In summary, we found evidence of a deformation mechanism transition at a bilayer thickness of 8 nm using the nanoindentation creep test. The high creep rates observed in the Ag/Ni multilayers with a small bilayer thickness reveal that the unusual mechanical behavior is attributable to grain boundary deformation. These results and analysis demonstrate that the crossover in the mechanical behavior of the multilayers is accompanied by a change in the underlying deformation mechanism, from a dislocation-dominated process to a grain boundary-dominated process.

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