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Strength of Ag-Ni multi-layered foils

J. ECHIGOYA*, C. NAKATA, J. TAKAHASHI, K. NONAKA, WULIN SONG
Department of Materials Science and Engineering, Faculty of Engineering, Iwate University,
4-3-5 Ueda, Morioka, 020-8551 Japan
E-mail: echigoya@iwate-u.ac.jp

The effect on mechanical properties was investigated of controlled variations in the layer thickness in multi-layered Ag-Ni foils. Multi-layered foils were made by hot pressing and then cold rolling of stacked foils. The thickness of a layer of multi-layers was controlled by the rolling rate. Tensile strength of foils increased with the decrease of thickness of a layer in the manner of the Hall-Petch relation up to a certain point and then maintained a constant value even when the thickness was further decreased. The effect of the thickness ratio of the two components on tensile strength was also studied in the Ag-Ni system.

1. Introduction

The mechanical properties of fine scale microstructures have received considerable attention in recent years [1]. It is known that a multi-layered system with thin layers results in an increase of density of the interface, which may result in behavior different from that of the bulk materials. Therefore, investigation of the strength of a layered structure consisting of thin films including metal–metal multi-layered materials is of great interest. The strength of thin multi-layered films and foils is mainly tested by the indentation method, which involves a complex deformation process, because of the restriction of sample size [2]. On the other hand, the tensile test yields information on flow stress and tensile strength, which can be interpreted from the dislocation motion. Koehler proposed the theory of strength enhancement of multi-layered metals and showed the possibility of designing strong materials in 1970 [3]. Lehoczky experimentally obtained the strength enhancement of thin multi-layered laminates proposed by Koehler [4]. The strength enhancement of multi-layers is mainly interpreted by the following two models: the Orowan-type model considering the motion of single dislocations proposed by Embury and Hirth, and the Hall-Petch model of dislocation pile-ups at the interface [5]. Reviews of this topic can be found in references [1, 2, 6]. A summary of various experimental results and measurements of mechanical properties of multi-layered films and foils performed by tensile and nano- or micro-indentation tests are presented in the table of Reference [2]. Electro-deposited Ni-Cu multi-layers with a thickness ratio of Ni:Cu = 9:1 have been examined by the tensile test, results showing that the yield stress and tensile strength peak at wavelengths (difference ratio of 1:1 ratio of Cu to Ni) of 40 and 60 nm, respectively [7, 8]. Experimentation performed by different investigators showed a peak in tensile strength at a wavelength of 20 nm and tensile strength which was also larger than that previously found [9]. Recently, Ag-Cu multi-layers with a layer thickness between 1.5 nm and 1.5 μm have been tested, results showing that the yield stress increased with decreasing layer thickness according to the Hall-Petch relation [10]. This $1/h^{1/2}$ dependence of the hardness of samples of Cu/metal multi-layered systems deposited by the sputtering method is evident in the so-called Hall-Petch plot obtained by Misra *et al.*, which shows that hardness increased with the decrease of layer thickness and became saturated of a thickness of less than about 50 nm in Cu-metal systems [10].

Though experimental studies have been reported, the measurement technique used to evaluate the mechanical properties has been mainly limited to the indentation test because of limitations of the common sample preparation methods, i.e., physical vapor deposition and electrodeposition [2]. Indentation is used to measure the multi-layered film on the substrate, but efforts to usea tensile test, which may give more direct information, have been few.

In the present study, the microstructure and mechanical properties of Ag-Ni foils with multi-layered structure made by hot pressing and then cold rolling were examined. Ag-Ni multi-layered samples with different thickness ratios of Ni and Ag were investigated and results were compared with those for the foils having layers of equal thickness.

2. Experimental procedure

Thin foils of silver and nickel or copper (99.9% purity and 0.01 mm thickness) were stacked and then hot pressed at 900 K for 1.8 ks under a stress of 20 MPa in a vacuum. The hot-pressed samples were rolled at room temperature. The layer thickness of the multi-layered foil was estimated from the reduction of samples after rolling. The samples used in the present experiment

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*Author to whom all correspondence should be addressed.

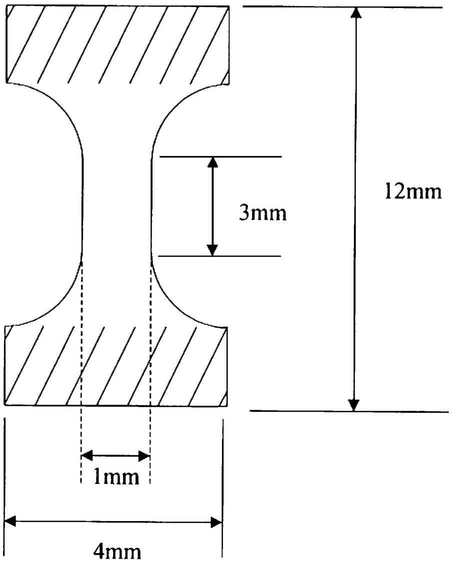

Figure 1 Specimen for tensile test.

had a strong preferred orientation of (110). Increasing the rolling reduction increased the degree of the preferred orientation. Samples with different thickness ratios of Ag-Ni multi-layered foil were prepared using foils with different starting thicknesses of 0.05 mm Ni and 0.01 mm Ag. The procedure was the same as that for the equal-thickness samples. The silver-nickel system is a eutectic system, in which the two components show little mutual solubility [12]. The structure after the hot pressing and rolling was examined by optical and transmission electron microscopy. Specimens for electron microscopic observation were prepared by ion milling. Transmission electron microscopy was performed with a Philips Tecnai 30. The tensile test was performed using an Instron-type tensile test machine with a strain rate of 0.05 mm/min. Specimens for tensile testing were cut out by a spark cutter, then polished with 3000 grid paper to remove damage due to the spark cutting process. The size and form of the specimens for the tensile test are shown in Fig. 1.

3. Experimental results

When the starting thickness of the foils was an equal thickness of 0.01 mm for both the Ag and Ni foils, the thickness ratio was 1:1. Fig. 2 shows a scanning electron micrograph of an Ag-Ni sample after hot pressing. A layered structure is produced and the interface is flat. Hot-pressed Ag-Ni was rolled to reduce thickness of the layers forming a multi-layered sample, and the thickness decreased with maintenance of the layered structure as shown in Fig. 3. Although layers were too thin to resolve each layer clearly, the structure seems to be a layered structure with a wavy interface.

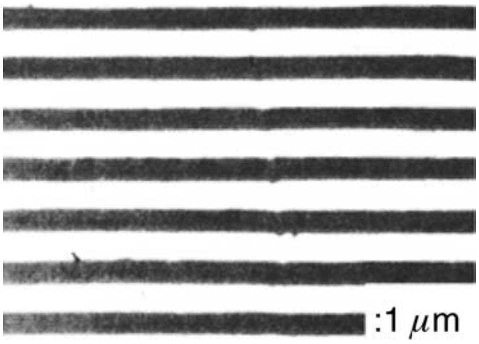

Figure 2 SEM micrograph of Ag-Ni (1:1) sample after hot pressing.

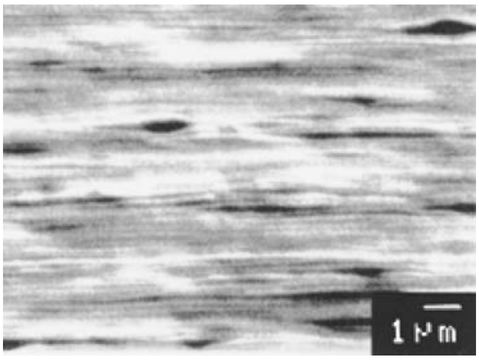

Figure 3 SEM micrograph of cold-rolled Ag-Ni (1:1) sample. h = 44 nm.

The stress strain curves of Ag-Ni multi-layered samples for different layer thicknesses are shown in Fig. 4. The foils were deformed plastically with 10% elongation. The stress strain curves of nickel and silver sheets, 0.01 mm in thickness are also shown in the figure. In comparison with these curves, the stress of the multi-layered sample is 3 times larger than that of the Ni and Ag sheets. This means that the rule of mixture of yield stress does not hold in the multi-layered system. Yield stress, defined as 0.2% proof stress, and tensile strength increased with decreasing layer thickness as seen in Fig. 4, which shows Ag-Ni multi-layered foils. From these figures, we can obtain the thickness dependence of the yield stress and the tensile strength for Ag-Ni(1:1) multi-layered foils as shown in Fig. 5. As the horizontal axis is taken by $1/h^{1/2}$, a linear

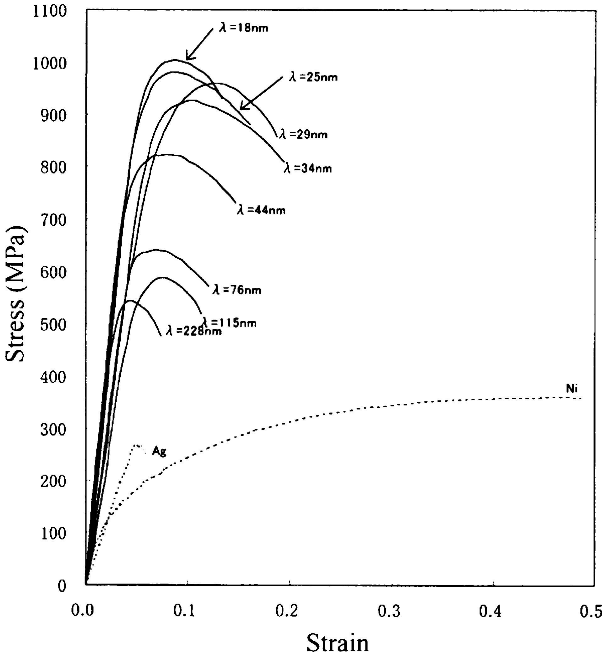

Figure 4 Stress strain curves of Ag-Ni (1:1) multi-layered foils.

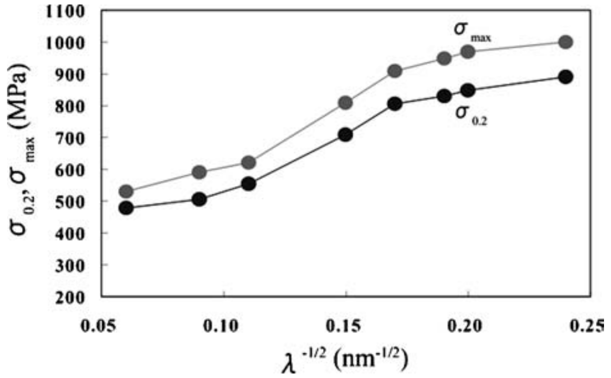

Figure 5 Dependence on 1/(thickness)$^{1/2}$ of yield stress and tensile strength of Ag-Ni (1:1) multi-layers.

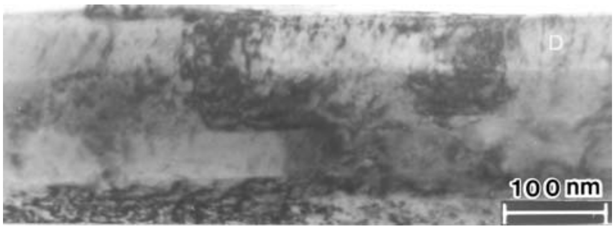

Figure 6 Transmission electron micrograph of a cross-sectional view of an equal-thick Ag-Ni multi-layered foil. $h = 72.5$ nm.

correlation indicates Hall-Petch behavior. The yield stress and the tensile strength of Ag-Ni multi-layered foils showed Hall-Petch behavior down to a layer thickness of around 25 nm, but this relation was no longer observed at layer thicknesses less than 25 nm, at which point the yield stress and the tensile strength became almost constant. Since dislocations were introduced by rolling, the parameter characterizing mechanical properties was selected to be the tensile strength in the present experiment. Though the foils were rolled to reduce the layer thickness, the thickness dependence of the yield stress, $\sigma_{0.2}$, and the tensile strength, $\sigma_{\max}$, had almost the same tendency. It should be noted that the fracture surface of the Ag-Ni multi-layered foils exhibited a dimple structure, which appears in ductile fractures. Fig. 6 shows a transmission electron micrograph of a cross-sectional view of a deformed equal-thick Ag-Ni multi-layered foil with a 72-nm-thick layer. The distribution of dislocations seems to be inhomogeneous, i.e., a multi-layer consists of layers with high dislocation density and layers with low dislocation density without any heavily tangled dislocations. The dislocations are parallel to the interface in some areas, but in other areas they are perpendicular to the interface. Dislocations, marked 'D' in Fig. 6, are across the layer but not elongated to the adjacent layer. The grain size in the layer seems to be much larger than the thickness of the layer. The interface between them is well defined and wavy.

Fig. 7 shows the stress strain curves of Ag-Ni (thickness ratio = 5:1) multi-layered foils. The curves are the same as those of Ag-Ni (1:1) multi-layered foils shown in Fig. 4, in which the decrease of the thickness led to the increase of the tensile strength and the decrease of elongation. The fracture mode was also ductile with dimples on the surface. The λ$^{-1/2}$ dependence of tensile strength of Ag-Ni (1:5) multi-layered foils is shown in Fig. 8, in which the dependence of Ag-Ni (1:1) foils was also indicated. Because the thickness ratio of Ag and that of Ni were different, a bilayer repeat length, λ, is used in the figure. The Hall-Petch relation was also satisfied over a wide range of λ. Though the shape of both curves is similar, the Ag-Ni (1:5) foils had higher stress than the Ag-Ni (1:1) foils for the same λ.

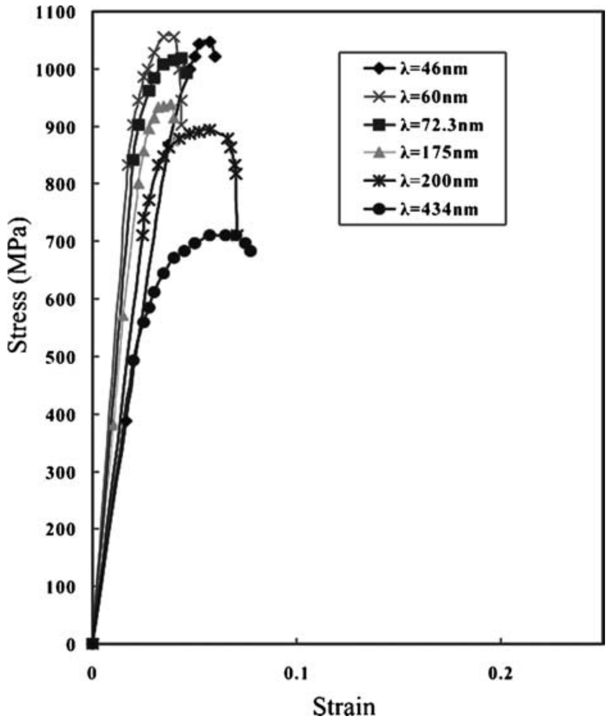

Figure 7 Stress strain curves of Ag-Ni (thickness ratio = 5:1) multi-layered foils.

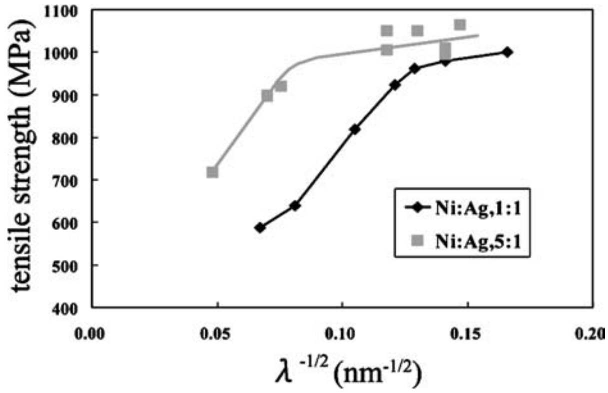

Figure 8 λ$^{-1/2}$ dependence of tensile strength of Ag-Ni (1:5) multi-layered foils.

4. Discussion

In polycrystalline materials, the grain boundary acts as resistance to the dislocation motion due to a pile-up of dislocations. The yield stress increment is inversely proportional to the square root of the grain size, d and is given by

$$\sigma = \frac{k_{\mathrm{HP}}}{\sqrt{d}}, \tag{1}$$

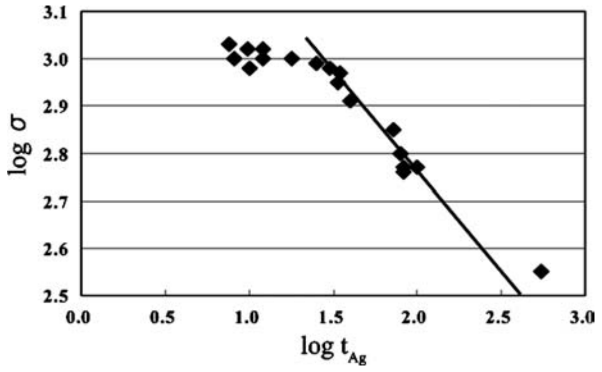

Figure 9 A log–log plot of tensile strength vs thickness of the Ag layer of Ag-Ni (1:1), (1:5) and (5:1) multi-layered foils.

where k_{HP} is a constant. If we apply this model to thin-layered materials regarding the grain size as the thickness of a layer, assuming that the yield stress and the tensile strength of thin layers will be dominated by the thickness effect, the yield stress will be inversely proportional to the square root of layer thickness. Since the Hall-Petch model is related to the stress of pile-up dislocations in the softer layer resulting in movement across the interface, it is necessary to use the layer thickness of Ag, t_{Ag}, rather than the bilayer repeat length of λ. Fig. 9 shows a log-log plot of $\sigma_{\max}$ vs. t_{Ag}, which allows comparison of the data obtained for foils of Ag-Ni (1:1), (1:5) and (5:1). The curve of a log-log plot becomes one curve, though the data used here are taken from the results of the different thickness ratios of Ag-Ni. If λ is employed as a parameter of the thickness, the thickness dependence of tensile strength shows two different curves as shown in Fig. 8. The gradient of the curve of part of the straight line for the large t_{Ag} region was almost 0.5, i.e., the Hall-Petch relation holds in this region. On the other hand, the Hall-Petch relation does not hold for a region of t_{Ag} smaller than 25 nm. A plot of the tensile strength of Ag-Ni (1:1), (1:5) and (5:1) multi-layered foils vs the inverse square root of the layer thickness of Ag gives $k_{\mathrm{HP}} = 0.125$ MPa $\sqrt{m}$ as shown in Fig. 10. The present experimental results revealed that the strength of Ag-Ni multi-layered foils was not affected by the thickness of the Ni layer.

The yield stress and tensile strength for all multilayers can be interpreted from the Hall-Petch model of Equation 1. When the thickness of a layer is smaller than 25 nm, a deviation from the Hall-Petch model occurs and a weak dependence or a strength plateau appears. The critical thickness for formation of the strength plateau depends on the thickness.

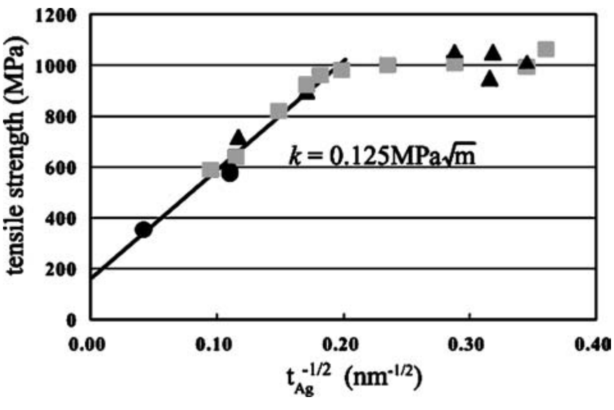

Figure 10 Tensile strength of Ag-Ni (1:1), (1:5) and (5:1) multi-layered foils vs the inverse square root of layer thickness of Ag.

A model explaining the deviation from the Hall-Petch relation has been given by Embury and Hirth [5]. When the dislocation is pinned at the interface, the interface dislocations are pure edge so that the moving segments are screw in character. In such a case, the Orowan stress necessary to move the dislocation segments is

$$\tau = \left[\frac{\mu b}{2\pi h} \ln\left(\frac{h}{b}\right)\right] \cos\phi, \tag{2}$$

where the core cutoff radius is set equal to Burgers vector, b. The angle is determined by a balance between the line tension of the moving screw segment and that of the edge embedded in the interface array. The deposition of interface dislocations leads to load transfer to the harder metal. The stress concentration in the harder metal causes flow to be initiated there. This model predicts that the initial yield stress is proportional to (ln h/b)/h. However, the strength increases with decreasing thickness, because (ln h/b) is larger than 1 for $h > 3b$. The condition of $h < 3b$, i.e., only the core of a dislocation exists in the layer, is not realistic.

Recently, Phillips *et al.* [13] proposed a model to explain the thickness dependence of strength of multi-layered films, but the fundamental idea is based on the Orowan model, which shows the increase of strength with decreasing thickness.

Arzt [6] proposed the following equation:

$$\sigma \approx \frac{\mu b}{\sqrt{d}} \ln \frac{d}{r_0}, \tag{3}$$

where r_0 is the core cutoff radius [6]. This equation can explain the decrease of strength with decreasing thickness smaller than critical thickness. However, the theoretical basis does not seem to be clear. In a thin layer, the strain energy of a dislocation decreases due to the factor of $\ln(h/r_0)$, which results in a decrease of the line tension of the dislocation and may facilitate dislocation movement [14, 15].

When the layer thickness is reduced, the number of pile-up dislocations becomes very small. At the limit where the transfer of slip across interfaces is left to single dislocations, the applied stress must be sufficiently large to supply the force needed to accomplish the transfer. Unlike pile-up theories of the Hall-Petch grain boundary strengthening, there are several models which explain the formation of a strength plateau when the thickness of a layer is smaller than the critical thickness [3, 16]. Koehler [3] proposed the image model in which the source of the increased resistance to dislocation motion is the shear modulus difference across the interface which results in image forces on the dislocation. Hoagland *et al.* [17] pointed out the importance of the alternating compression-to-tension in plane coherency strains which enable matching of the lattice parameters in the two adjacent lattices. Since the interface barrier to direct slip transmission given by these models is independent of layer thickness, these mechanisms should be the main factors resulting in a strength plateau.

5. Conclusion

We have studied the variation in tensile strength of multi-layered Ag-Ni foils caused both by varying the absolute layer thicknesses and by varying the ratios between these thicknesses as 1:1, 1:5 and 5:1. The samples were prepared by hot pressing and then cold rolling stacks of foils. The thickness of the resulting stack of multi-layers was controlled by the rolling rate. Tensile strength of foils increased with the decrease of thickness of an Ag layer in the manner of the Hall-Petch relation up to a certain point and then maintained a constant value even when the thickness was further decreased.

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