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Grain-size effect on fatigue properties of nanocrystalline nickel thin films made by electrodeposition

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Abstract

Nickel nanocrystalline thin films were produced by electrodeposition using sulfamate solution. Free-standing thin films with various grain sizes ranging from 9 to 67 nm were produced under a constant current with changing the content of brightener and temperature of electrodeposition. The grain-size effect on the fracture strength and the yield strength in tension tests can be divided into two regions: region A with the grain size larger than about 15 nm and region B with the grain size between 9 to 15 nm. In region A, the tensile strength and yield strength increase following Hall-Petch relation, and the tensile elongation decreases with decreasing grain size. In region B, the tensile and yield strength increases following a different relation of Hall-Petch type, and the tensile elongation also increases with decreasing grain size. The fatigue strength also increases with decreasing grain size following different Hall-Petch relations between region A and B. The resistance to fatigue crack propagation decreases for nanocrystalline films. Scanning electron microscopic observation of deformation near cracks shows two different features depending on the grain-size region A or B. In region A, ordinary slip deformation within the grain is predominant, while grain boundary sliding is more important in region B for fatigue damage formation.

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Keywords: Fatigue; nickel nanaocrystals; thin films; grain-size effect; Hall-Petch relation; crack propagation; fractography

1. Introduction

Nanocrystallization is one of the most promising technique to improve the fatigue strength of metallic thin films which are now being widely used for micro-electro-mechanical systems. The electrodeposition method will be the easiest method to obtain a uniform nanocrystalline structure in thin films [1,2]. In

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comparison with hardness and tensile properties, fatigue properties of nanocrystalline metals are not well understood. Ultrafine-grained copper made by equal-channel angular processing (ECAP) has a high fatigue resistance at an intermediate life region, but the improvement of the fatigue strength diminishes near the fatigue limit because of grain growth by cyclic deformation [3]. Since nickel may not show grain growth by cyclic deformation during fatigue, the improvement of the fatigue strength due to nanocrystallization remains even near the fatigue threshold [4,5].

In the present paper, nickel nanocrystalline thin films were produced by electrodeposition using sulfamate solution. Various thin films with different grain sizes ranging from submicrometer to nanometer were produced by changing the elecrodeposition conditions. The grain-size effect on the tensile and fatigue properties was investigated. Fractures surfaces were examined by scanning electron microscopy (SEM) to understand micromechanisms of fracture.

2. Electrodeposition of Nickel Thin Films

Nickel thin films were produced by electrodeposition using sulfamate solution. A polished stainless steel plate was used for a cathode and a pure nickel plate for an anode. The amount of brightener additive was changed at six levels from 0 to 2 g/L. The temperature of the bath was kept constant at 313 or 328 K in a hot-water circulating bath. The pH value of the solution was maintained at 3.7 to 4.2 by adding sulfamate acid. The solution was stirred by a magnetic stirrer to avoid pit formation. The current density was kept constant at 25 mA/cm^2 by using a constant current supply, and the deposition period was 28 min. The thickness of the thin film was around 10 μm.

After deposition, thin films were removed from the cathode and subjected to the characterization of the tensile and fatigue properties as free-standing films. The microstructure of thin films was examined by X-ray diffraction, scanning electron microscopy (SEM), electron backscatter diffraction (EBSD) and transmission electron microscopy (TEM).

The grain size of films was determined by X-ray line broadening of 200 diffraction. The diffraction profile was approximated by Cauchy function and the grain size was determined by the full-width at half maximum [6]. Table 1 summarizes the grain size determined X-rays. The addition of brightener quickly reduced the grain size from around 100 nm to 10 nm. The lower temperature of deposition gave a smaller grain size. The smallest is 9.0 nm of CC-ally (2g/L,313K) film, named NC film. The X-ray value agrees well with TEM observation showing a fairly uniform distribution of the grain size with the average of 15 nm [5]. The thin films made without brightener at 328K has a large grain size and is named here SM film. EBSD observation shows the grain size of 670nm which is larger than the value determined by X-rays [5]. The texture of thin films was examined by X-ray diffraction. Films with small grain sizes show random orientation, while films with larger grain sizes have a strong 200 fiber texture. The grain orientation of each film is also shown in Table 1, where the boundary is around 15nm in grain size.

Temperature	CC	CCally (0.25g/L)	CCally (0.5/L)	CCally (1.0g/L)	CCally (1.5g/L)	CCally (2g/L)
328 K (55°C)	67.7 nm*	35.2nm	16.4 nm	14.1nm	12.0 nm	12.2 nm
313 K (40°C)	141.9 nm	21.3 nm	12.9 nm	10.3 nm	9.4 nm	9.0 nm**
	← 200 texture →		← Random orientation →			

* CC(328K) film is named SM film
** CCally(2g/L,313K) film is named NC film

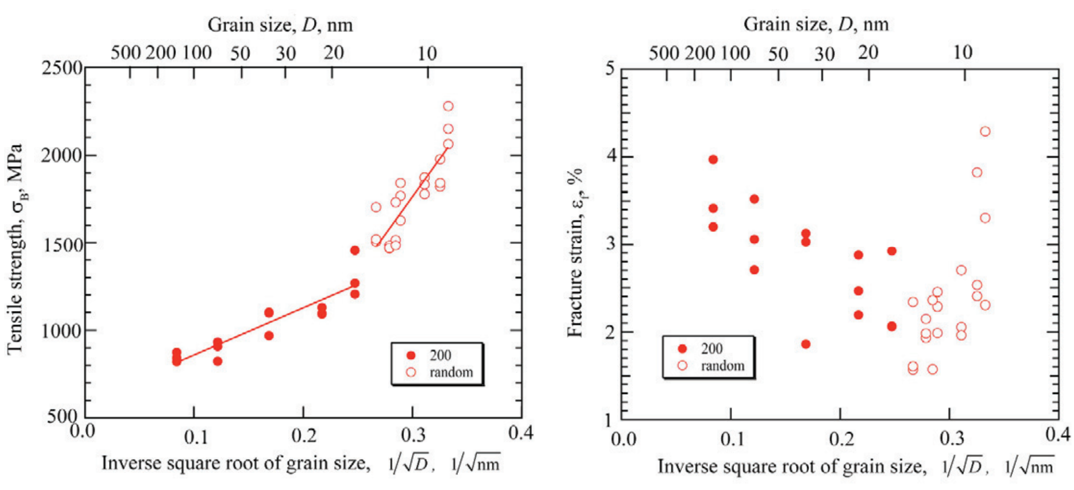
Table 1. Grain size determined by X-ray diffraction.

3. Tensile Properties

Tensile tests were conducted using dumbbell type specimens with a width of 4 mm and a gage length of 10 mm. The thickness was adjusted to have 10 μm and the exact thickness was determined from the fracture surface by SEM. For each thin film, three specimens stretched at the crosshead speed of 0.1 mm/min, and the elongation was measured by a laser dimension measurement equipment (KEYENCE, LS-7600, LS-7030M).

The tensile fracture took place at the maximum load without any load drop. Figure 1(a) shows the relation between the tensile strength and the inverse square root of the grain size, and Fig. 1(b) shows the change of the elongation with the grain size. The tensile strength and the 0.2% proof stress increased with the grain size. It is interesting to note that the elongation tends to decrease with decreasing grain size down to about 15nm, and then increases with decreasing grain size. The grain-size effect on the tensile properties can be divided into two regions: region A with the grain size larger than about 15 nm and region B with the grain size between 9 to 15 nm. In region A, the tensile strength and yield strength increase following Hall-Petch relation as in ordinary grain-sized metals. Dislocation motion within grains is the predominant mechanism of plastic deformation. The strength increase caused by impeding dislocation motion results in the lower ductility by dislocation accumulation. In region B, both strength and ductility increase with decreasing grain size. There is no dislocation accumulation as revealed by a reversible broadening of the X-ray line profile during loading and unloading. Grain boundary sliding may be responsible for enhanced ductility.

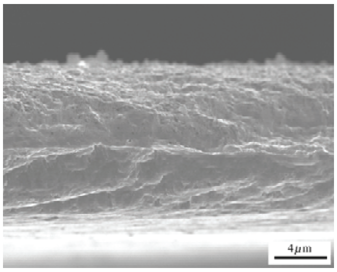
The topography of tensile fracture surfaces is distinctly different between region A and B. Figure 2 (a) shows a chisel edge fracture following necking in thin film grouped into region A, while Fig. 2(b) shows flat fracture surface without necking in thin films of region B. enhanced ductility.



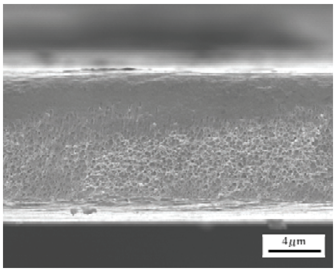
(a) Tensile strength

(b) Fracture strain

Fig. 1. Effect of grain size on tensile properties.



(a) SM film, CC(328K)



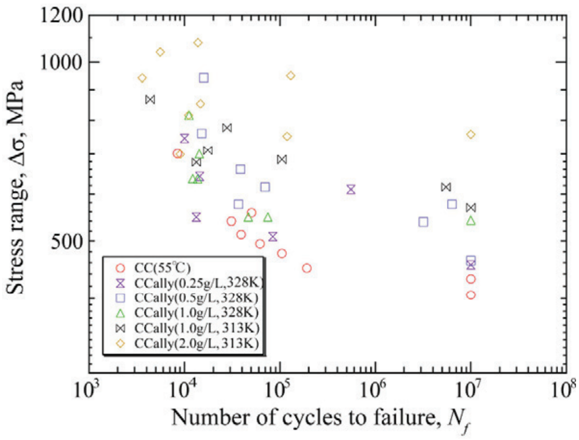
(b) NC film, CCally (2g/L,313K)

Fig. 2. SEM micrographs of tensile fracture surface.

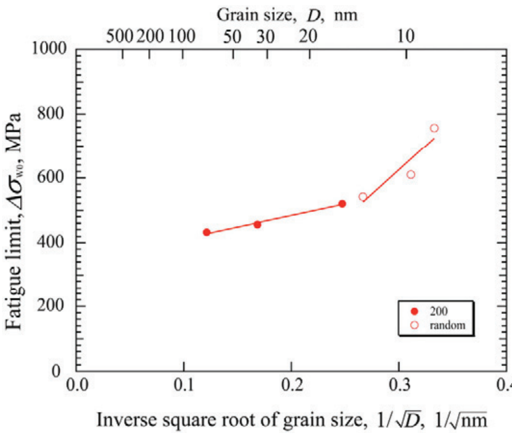
4. Fatigue Strength

Dumbbell-type specimens were machined from free-standing thin films which has thickness of 10 μm and a width of 2 mm. Fatigue tests were conducted in a servo-electromagnetic fatigue testing machine (Shimadzu MMT-100N-10) under the stress ratio of R=0.1 at a frequency of 20 Hz.

The relation between the applied stress range and the number of cycles to failure is shown in Fig. 3(a) for six thin films. The fatigue strength of thin films is much larger than the bulk nickel, and increases with decreasing grain size. The fatigue limit corresponding to 10^7 cycles of NC films reaches to 757 MPa and is about 2 times larger than that reported by Hanlon et al. [4] for nanocrsytalline nickel of 20-40 nm grain size. Not only grain size but also some other microstructural feature is expected to be responsible for the improvement of the fatigue strength. The fatigue limit is plotted against the inverse square root of the grain size in Fig. 3(b). Hall-Petch relation is satisfied in region A and B, but the relations are different between region A and B. The slope of Hall-Petch relation is higher in region B than in region A. Together with a large improvement near the fatigue threshold, even at an intermediate life region, some improvement of fatigue strength is seen in Fig. 3(a). This shows a clear contrast to the fatigue strength of ECAPed copper whose improvement near the fatigue threshold disappears because of grain growth due to cyclic deformation [3]



(a) S-N relation



(b) Hall-Petch relation.

Fig. 3 S-N relation and Hall-Petch relation for fatigue limit.

5. Fatigue Crack Propagation Behavior

Fatigue crack propagation tests were conducted using single edge notched films with 5 mm in width and 10 μm in thickness under a stress ratio of the minimum to maximum stress R = 0.1. The stress intensity factor was calculated by the modified crack-closure integral method of the finite element method under the experimental boundary condition of gripping. The near-threshold crack propagation behavior was determined by the load-shedding method.

Figure 4 shows the relation between the crack propagation rate and the stress intensity range for four thin films. The resistance to fatigue crack propagation is reduced for nanocrystalline CC-ally films, and the threshold stress intensity factor of these materials were lower than that of SM films.

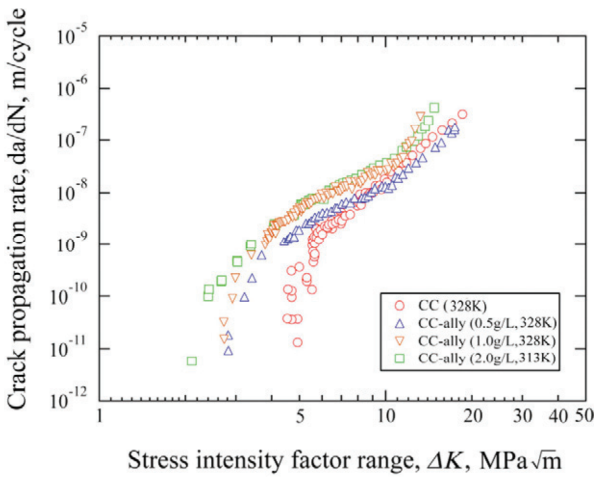


Fig. 4 Relation between crack propagation rate and stress intensity range.

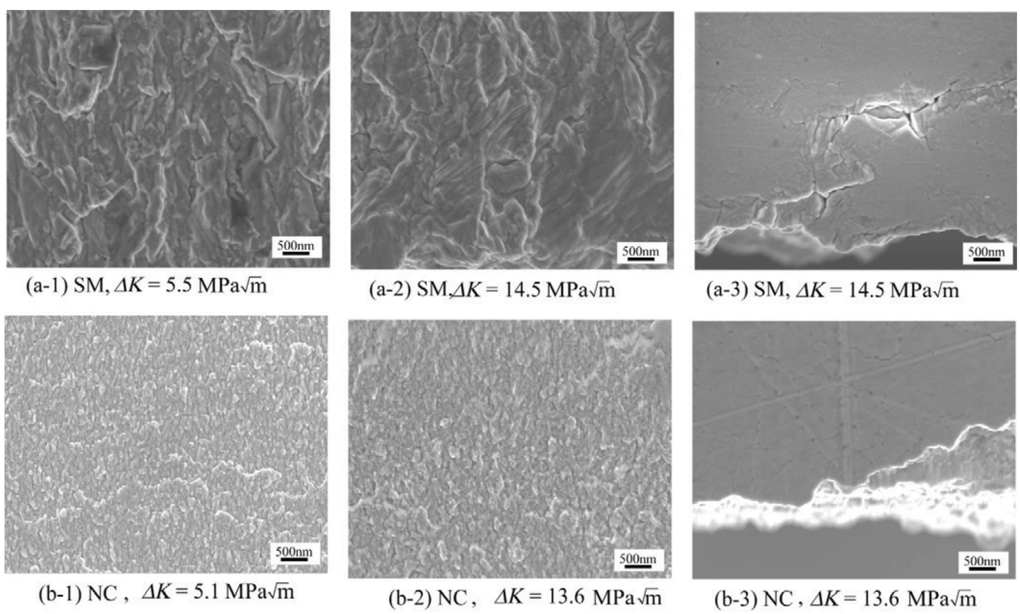


Fig. 5 SEM micrographs of fatigue fracture surface of SM and NC films.

Figures 5 show SEM micrographs of the fracture surfaces (left column) and the film surface near the fracture line (right column) for SM and NC films. The crack propagation direction is from left to right. Granular features are seen on the fracture surface near the threshold in NC films for both low and high stress intensity ranges. At high stress intensity range, striations are detected for SM films. A granular feature seen near the threshold is similar to the feature found in NC films. On the specimen surface near the fracture surface at high stress intensities, slip deformations are evident for SM film. There is not trace of slip deformation of grain boundary fracture for SM film. The roughness of the fracture surface is low for NC films compared with SM films. A small amount of roughness may result in a smaller amount of crack closure, and consequently less amount of resistance to crack propagation. The crack propagation mechanisms for SM films are not different from ordinary microcrystalline materials, while other mechanisms related to grain boundary will be operating for nanocrystalline materials.

6. Conclusions

Nickel nanocrystalline thin films were produced by electrodeposition using sulfamate solution. Free-standing thin films with various grain sizes ranging from 9 to 67 nm were produced under a constant current with changing the content of brightener and temperature of electrodeposition. The grain-size effect on the fracture strength and the yield strength in tension tests can be divided into two regions: region A with the grain size larger than about 15 nm and region B with the grain size between 9 to 15 nm. In region A, the tensile strength and yield strength increase following Hall-Petch relation, and the tensile elongation decreases with decreasing grain size. In region B, the tensile and yield strength increases following a different relation of Hall-Petch type, and the tensile elongation also increases with decreasing grain size. The fatigue strength also increases with decreasing grain size following different Hall-Petch relations between region A and B. The resistance to fatigue crack propagation decreases for nanocrystalline films. Scanning electron microscopic observation shows two different features depending on the grain-size region A or B. In region A, ordinary slip deformation within the grain is predominant, while grain boundary sliding is more important in region B.

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