Texture and Grain Growth in Nanocrystalline Permalloy

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ABSTRACT

The texture evolution accompanied by grain growth was investigated in a nanocrystalline Fe-78wt%Ni alloy, namely permalloy fabricated by using an electrodeposition method. The as-deposited specimen revealed a strong <100>//ND and a weak <111>//ND fibre-type textures. Grain growth took place during annealing beyond 370°C and resulted in the texture change such that the <111>//ND fibre strongly developed at the expense of the <100>//ND fibre. It was observed using orientation image microscopy (OIM) that the <111>//ND oriented grains abnormally grew in the early stages of grain growth. The relationship between the texture evolution and the microstructural change during annealing is interpreted and discussed in terms of the orientation dependency of grain growth

Keywords : electrodeposition, grain growth, nanocrystalline, permalloy, texture.

1 INTRODUCTION

Nanocrystalline materials consisting of nanometer-sized crystallites contain a large number of interfaces, and thus, a large volume fraction of the atoms are associated with the intercrystalline region [1]. Due to this morphological characteristic, in general, nanocrystalline materials are far away from thermodynamic equilibrium and as a result reveal quite different properties in comparison with conventional coarse-grained materials [2].

For example, in ferromagnetic materials, the soft magnetic properties can be significantly modified by reducing grain size to nanometer scale [3-5]. In fact, a particular attention has been paid to the synthesis of nanocrystalline materials for better soft magnetic properties. As an attempt for these materials, extensive studies [6-7] on structure-property correlations in nanocrystalline Ni-Fe alloys have been carried out in recent years.

In the present work, a nanocrystalline Fe-78wt%Fe alloy was fabricated using an electrodeposition method. Fe-Ni alloys exhibit different physical properties with different chemical compositions, among which Fe-78%Ni alloy, referred to as permalloy, has widely been used owing to an outstanding combination of soft magnetic properties. Such nanocrystalline electrodeposits are in a high non-equilibrium (metastable) state, i.e. a high energy state due to a large number of interfaces [1] resulting in the occurrence of grain growth at relatively lower temperatures.

The present study was aimed at investigating grain growth that takes place during the annealing of the nanocrystalline Fe-78%Ni alloy electrodeposits. In particular, early stages of grain growth were examined with the aid of orientation image microscopy (OIM) techniques and discussed in terms of the evolution of textures.

2 EXPERIMENTAL PROCEDURE

A nanocrystalline Fe-78%Ni alloy foil was fabricated by using an electrodeposition method in a modified Watts-type bath containing nickel chloride, iron sulfate, boric acid and saccharine. Deposition was carried out at a temperature of 45°C and at a current density of 10 A/dm^2. The final thickness of the electrodeposited foil was approximately 20 μm. The grain size of the electrodeposit was calculated to be 10-20 nm using the Scherrer formula [8] on the basis of the X-ray diffraction (XRD) peak broadening.

Thermal behavior of the specimens was examined using a DSC, where the samples were heated from room temperature up to 450°C at a heating rate of 10°C/min. On the basis of the DSC data, the specimens were annealed at temperatures ranging from 250°C up to 400°C for different heating times in an infrared heating furnace in an argon atmosphere.

X-ray pole figures were measured using Co K_α to determine the development of macrotextures. For the analysis of microtextures, the samples were observed in a field emission gun scanning electron microscope (FEGSEM) with an automated orientation imaging microscopy (OIM) system. OIM scans were carried out in a hexagonal grid of 20 nm step size.

3 RESULTS

Figure 1 shows typical DSC curves obtained during heating of the Ni-base alloys with different contents of Fe fabricated in the current work. The exothermal processes

are indicated by the appearance of heat release peaks. The temperature at which the exothermal process takes place

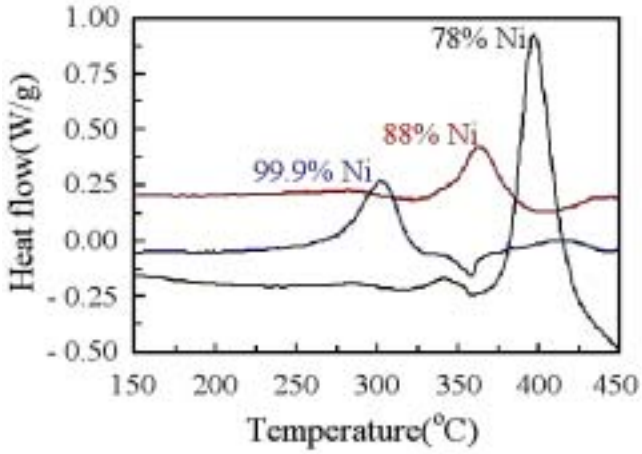


Figure 1: DSC curves obtained during heating nanocrystalline Ni-Fe alloy electrodeposits.

decreased with increasing Ni content. These exothermal processes are attributed to the occurrence of grain growth in the samples.

Grain growth occurs in polycrystalline materials to decrease the interfacial energy and hence the total energy of the system. Since nanocrystalline materials are in a high energy state due to a large volume fraction of the interfacial component, the driving force for grain growth is so large that the grain boundaries can readily move far below temperatures at which grain growth is expected to occur in microcrystalline materials. It is estimated that the heat energy released during grain growth in a 5nm-sized material is about 2-3 orders of magnitude larger than that in a 5μm-sized material [9].

Klement et al. reported that grain growth occurs in the temperature range between 353K and 562K in electrodeposited nanocrystalline pure Ni [10]. Their result is well fitted to the DSC curve of 99.9%Ni as shown in Figure 1. In the DSC curve for the Fe-78%Ni alloy foil, the heat release peak starts at 373.1°C, i.e. the onset temperature and reaches the peak maximum at 391.4°C.

Figure 2 shows the XRD scans to examine the occurrence of grain growth in this temperature range. In the samples annealed for 5 min up to 350°C, the {111} diffraction patterns are similar to that of the as-deposited sample with regard to XRD peak intensity and broadening. The small changes of the peak shape can be attributed to the transformation of the highly disordered interfacial component into a lower energy state without changing the grain size. However, in the sample annealed at 400°C for 5 min, the peak intensity drastically increased and the full width at half maximum of the peak significantly decreased. The latter indicates the occurrence of grain growth in the temperature range between 350 and 400°C.

Grain growth is accompanied not merely by the change in dimension but also by a structural rearrangement, i.e. a change of crystallographic textures.

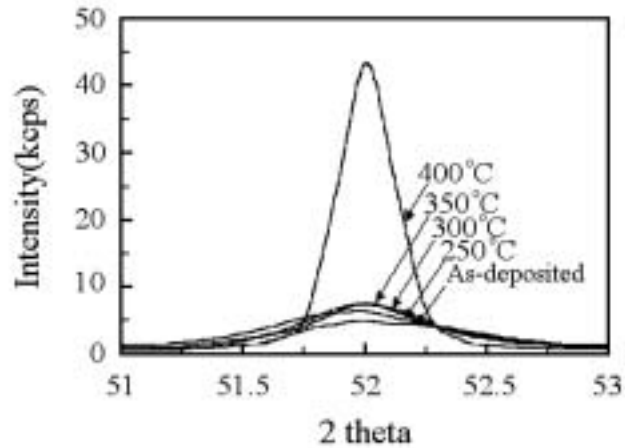


Figure 2: Change of {111} XRD peaks in the specimens annealed at different temperatures.

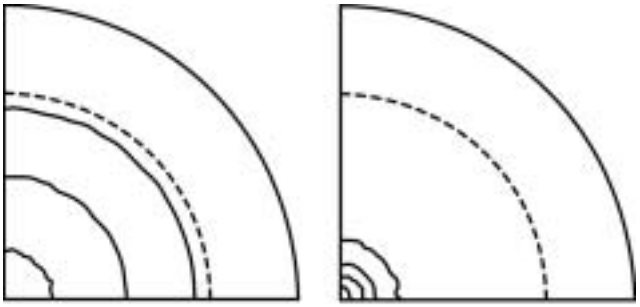
Figure 3 depicts the {111} and {100} pole figures in the as-deposited specimen. The texture development is characterized by a mixture of a strong <100> parallel to the sheet normal direction (//ND) and a weak <111>//ND fibre components. This textural state was completely reversed after annealing. As shown in Figure 4, in the specimen annealed at 400°C for 5 min, the development of the <111>//ND fibre texture became predominant while the intensity of the <100> fibre texture decreased to a low level. Therefore, it is obvious that during annealing, the <111>//ND oriented grains grew faster than <100>//ND and other oriented grains.

4 DISCUSSION

Recently, Li, Czerwinski and Szpunar [11] calculated the orientation dependency of surface energy by using atomistic interactions by Lennard-Jones in electrodeposited Ni-Fe alloys. According to their calculation, the surface energy is the lowest for the {111} plane and increases in the order of {100}, {110}, {311}, {210}, etc. It follows that the <111>//ND texture is expected to strongly develop when the electrodeposition is carried out under a condition close to equilibrium. As the deposition condition becomes farther from equilibrium, crystallographic planes having higher surface energies, e.g. {100}, would increase rather than {111}.

Suppose two nanocrystalline electrodeposits consisting of only the <100>//ND and only the <111>//ND grains, respectively. It is apparent that the free energy of the {100}-grained material is higher than that of the {111}-grained material since the latter may be electrodeposited under a condition closer to equilibrium. There can exist two possibilities that the energy difference between the two materials results in microstructural difference: first, in case

that the two materials have the same grain size, energy per unit volume of the <100>//ND grains would be higher than

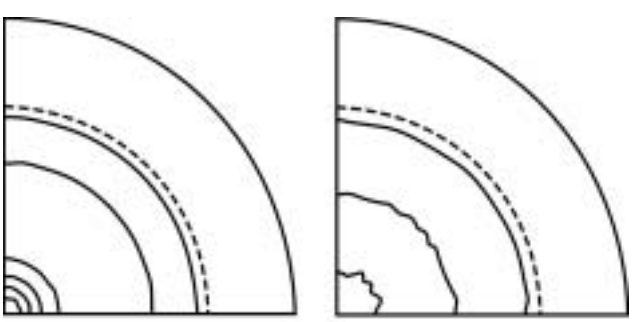


{111} {200}

Figure 3: {111} and {200} pole figures (intensity levels: 1, 2, 3 and 4; max=4.43) in the as-deposited Fe-78%Ni foil.

that of the <111>//ND grains; second, in case that grains with different orientations have the same energy density, the size of the <111>//ND grains would be larger than that of the <100>//ND grains.

These two possibilities will be now employed to explain the texture change that took place during the annealing of the current material. The as-deposited specimen consists of the <100>//ND grains as the major component and the <111>//ND grains as the minor component. Therefore, our material lies somewhere between the two extremities above, i.e. only-{100}-grained and only-{111}-grained materials. Under assumption that energy density is independent of crystallographic orientation, the <111>//ND grains whose size might be the largest in the material would grow faster due to the size advantage to dominate the annealing texture. However, from OIM observation of the as-deposited specimens, there is no evidence that the size of the <111>//ND grains is larger than that of other oriented grains. On the other hand, if grain size is the same regardless of the orientations of grains, the {111} grains having the lowest energy density may grow into the different oriented grains to decrease the total energy of the system.



{111} {200}

Fig. 4. {111} and {200} pole figures (intensity levels: 1, 2, 3 and 4; max=4.94) in the electrodeposited Fe-78%Ni foil annealed at 400°C for 30 min.

Figure 5: OIM map showing the abnormal growth of {111} grains after annealing at 380°C for 1 min.

In reality, the OIM map obtained in the sample annealed at 380°C for 1 min shows that the <111>//ND grains were rapidly grown to be much coarser than other oriented grains as in Figure 5. It is of importance that abnormal growth of the <111>//ND grains occurred in the early stages of grain growth. The driving force for abnormal grain growth is fundamentally the reduction in grain boundary energy as for normal grain growth. However, abnormal grain growth can occur only when normal grain growth is inhibited due to such factors as impurities, second-phase particles, textures, etc. [12].

Figure 6 shows the XRD patterns before and after annealing the electrodeposited Fe-78%Ni alloy foil. In the specimen annealed at 400°C for 30 min, when compared to the as-deposited specimen, the diffraction angle for the {111} peak decreased while that for the {200} peak increased. This indicates that the interatomic spacings of the <111>//ND grains increased and the <100>//ND grains shrank during annealing. Therefore, the abnormal growth of the <111>//ND grains might be attributed to their lower energy density as well as atomistic expansion in the early stages of grain growth.

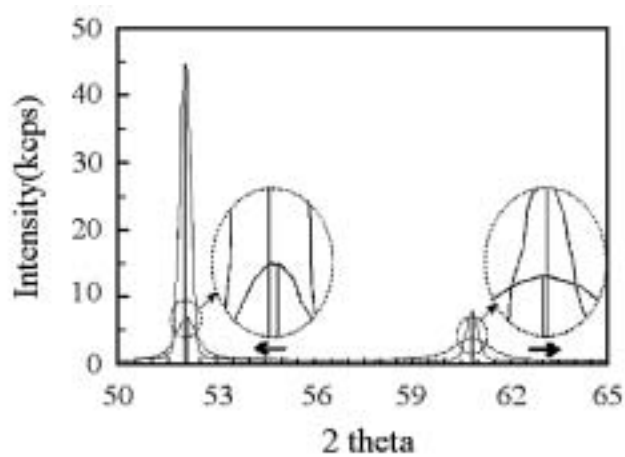


Figure 6: Difference of the {111} and {200} peak positions

in the electrodeposited Fe-78%Ni foil before and after annealing.

5 SUMMARY

Grain growth that takes place during the annealing of electrodeposited nanocrystalline Fe-78wt%Ni alloy foil was investigated and discussed in terms of its possible effect on the texture evolution. The texture of the as-deposited specimen was characterized by a strong <100>//ND and a weak <111>//ND fibres. Abnormal growth of the <111>//ND grains in the early stages of grain growth occurred on annealing beyond 370 °C and resulted in the texture change that the <111> fibre strongly developed at the expense of the <100> fibre. This texture evolution was interpreted by means of the orientation dependency of grain growth.

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