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Stress Induced Phase Transition of Iron-Rhodium Alloys

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Abstract. Stress-induced phase transitions (B2-L1₀, B2-fcc) on an FeRh alloy were investigated with X-ray diffraction (XRD) and transmission electron microscopy (TEM). An Fe-50.5at%Rh alloy was rolled to 80μm thickness, and annealed at 1370K for 173ks. Annealed sample sheets were cold rolled at various rolling rates, and changes of the sample alloy on the phase state were investigated with XRD. The L1₀ phase appeared in the early stage of cold work. With heavy work appearance of the fcc phase and consequent decrease of L1₀ were observed. Destruction of the order of B2 phase was also observed with progress of cold working. The crystallography and morphology of stress induced phases were studied with TEM. L1₀ phase grains were appeared as layers of about 100 to 1000nm thickness, and density of dislocations in them were extremely high. The separate appearance of fcc phase from L1₀ phase was found in B2 phase grains. The result is negative to an idea that the L1₀ phase is an intermediate phase of the B2-fcc transition.

1. Introduction

Iron-rhodium alloys containing Rh from about 20at% to 50at% have a B2 ordered structure (α₁ phase) at room temperature in thermal equilibrium.[1] The most interesting phenomenon that occurs in the Fe-Rh alloy system might be the first order magnetic phase transition of near-equiatom B2 ordered alloys between antiferromagnetic and ferromagnetic phases.[2,3] However, there are other peculiar phase transitions in this system; two types of stress-induced phase transition occur on the same B2 ordered alloys with cold working. An L1₀ structure appears from original B2 phase alloys with comparatively mild work, and with heavy work comes out a disordered fcc structure which is the thermal equilibrium phase at high temperature (γ phase).[1]

The transition to fcc structure was found rather earlier than the other stress-induced phase transition: It was first reported in 1960's by Lommel and Kouvel, who reported that fcc single phase powder was produced from B2 ordered FeRh alloys by filing at room temperature, and the same phase transition could be induced by cold rolling.[4] They observed the change in the magnetization of the stress induced fcc phase with temperature. The fcc phase was paramagnetic at room temperature, and metastable under 510K. At higher temperatures increase of magnetization was observed, which was associated with reverse-transition of fcc to B2. On the other hand, it was rather recent that the transition to L1₀ phase was found. Miyajima and Yuasa reported it first in 1992 in their paper on magnetic properties of FeRh_{1-x}Co_x. [5] They found out appearance of L1₀ phase in B2 ordered FeRh alloys under cold rolling, and the lattice constants of it for bct unit cell were 0.281nm and 0.335nm along the a and c axes, respectively. They have studied the relations between magnetic properties and crystal structures about FeRh_{1-x}M_x (M=Co, Pt, Pd) alloys, [5,6,7] and reported similar phase transitions that occurred on FeRh_{1-x}Pt_x (x<0.1) alloys which have a B2 structure as the stable phase.

These stress induced phase transitions occur at room temperature and are thought to be martensitic. However, there are very few studies on their nature; morphology, crystallographic relationships, behavior of progress, and so on. Aiming to solve these problems we performed cold rolling of an Fe-50.5at%Rh alloy and investigated the process of stress induced transformations of the B2 ordered phase in it by X-ray diffraction (XRD), and the morphologies by careful transmission microscopy (TEM). With TEM, we succeeded a direct observation of stress induced phases in the B2 phase grains, and crystallographic relationships between L1₀ and B2 phases were determined by electron diffraction.

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2. EXPERIMENTAL PROCEDURE

A specimen alloy of Fe-50.5at%Rh was induction melted and casted in a vacuum. The ingot was hot forged and rolled to 1mm thickness in air. Surface oxide layers grown in course of hot working were ground off mechanically and the ingot was homogenized at 1370K for 173ks in an evacuated silica tube. Sample sheets of 80 μ m thickness were prepared by cold rolling. In course of cold rolling, the sheets were annealed several times to avoid fracture. To remove effects of cold work, sample sheets were annealed at 1370K for 173ks again before experiments.

Annealed specimens were investigated with XRD firstly, and cold rolled at various rates so far as 75% to get stress induced phases. Rolled specimens were examined with XRD and TEM. X-ray diffraction was measured in the range of scattering angle 2θ from 30° to 150° using Co K α radiation. Specimens for TEM were prepared by Argon ion thinning. Disks of 3mm diameter were punched from rolled specimens. The center of a disk specimen was dimpled mechanically as preliminary rough thinning. Ion thinning was performed with Gatan Duomill model 600 operated at 2.5kV. Ion beam current was kept under 0.15mA to avoid unexpected superheating of the specimen. TEM observation was performed with JEOL JEM-200CX.

3. EXPERIMENTAL RESULTS

3.1. X-ray diffraction

The XRD patterns are shown for an as-quenched Fe-50.5at%Rh specimen and after 40% rolling in Fig.1(a) and (b), respectively. In the pattern of the as-quenched specimen strong reflections from B2(α_1) phase and rather weak reflections from fcc(γ) phase are seen. According to the conventionally used phase diagram, an alloy of this composition takes a B2 single phase in thermal equilibrium, and the fcc phase of the specimen might seem to be a trace of the high temperature phase. However, the fcc reflections never shrink after extremely long annealing at 1370K. Because of this fact and some other evidences obtained by our former work using the same alloy,[8,9] we believe the thermal equilibrium state of an Fe-50.5at%Rh alloy at 1370K is a mixture of B2 and fcc phases. In the pattern of the rolled specimen, there appear new reflections corresponding neither B2 nor fcc. They are from the stress induced L_{10} phase. Lattice parameters for the bct unit cell are 0.283nm and 0.333nm along the a and c axes, respectively.(c/a=1.18) Difference of these values from former report[5] may be caused by difference of compositions or heat treatment of specimens. Progress of the transition to fcc phase is shown by intensified fcc reflections.

The changes of XRD patterns with rolling rates are shown in Fig.2. At a rolling rate of 5%, there already appear reflections from L_{10} phase. Increase of L_{10} reflections continues up to a rolling rate of 30%. In contrast, reflections from fcc phase scarcely increase at small rolling rates. Obvious increase of fcc reflections is observed at a rolling rate of 30% for the first time. Once fcc reflections begin to increase, other reflections from the B2 and the L_{10} phases start to shrink. At a rolling rate of 75%, all distinct reflections are from fcc phase, and the reflections of other phases are only traces.

Intensity ratio of 100_{B2} order reflection to 200_{B2} fundamental reflection was calculated in order to evaluate destruction of the order of B2 phase with cold work. Results of calculation are plotted for the changes of rolling rates in Fig.3. The ratio shows prominent decrease with the progress of cold work.

3.2. Transmission electron microscopy

In Fig.4 are shown the internal structure of an as-quenched Fe-50.5at%Rh alloy from 1370K. In (a) of a low magnification image, spherical grains of about 1 μ m diameter are seen among matrix grains. These spherical grains have an fcc structure, and the matrix grains are B2. High magnification images are shown

Figure 3: The changes of $100_{B2}/200_{B2}$ intensity ratio with the strength of cold rolling.

Figure 1: XRD patterns from (a) an as-quenched Fe-50.5at%Rh specimen and (b) after 40% cold rolling.

Figure 2: XRD patterns from Fe-50.5at%Rh specimens cold rolled at various rolling rates: the rolling rate corresponding each pattern is shown above.

Figure 4: Internal structure of an as-quenched Fe-50.5at%Rh alloy. (a) Low magnification image: spherical fcc grains in a B2 grain are seen near the edge. Higher magnification of B2 and fcc grains are shown in (b) and (c), respectively.

Figure 5: $L1_0$ phase grains appeared in a B2 grain. (a) TEM micrograph and (c) SAD pattern taken from 110 pole of $L1_0$ grains, and (b) and (d) are taken from the 110 pole of the B2 grain.

Figure 6: Internal structure of a $L1_0$ grain and the SAD pattern (inset). The $L1_0$ grain is surrounded both sides by B2 grains.

in (b) and (c), and the micrograph (b) shows a B2 grain, and (c) does an fcc grain. Dislocations exist in the B2 grain but scarcely do in fcc, and sharp twin contrasts are seen in the fcc grains.

Stress induced phases are observed in specimens rolled at a rate of 30%. $L1_0$ grains appeared in a B2 grain and a SAD pattern from one of them are shown in Fig.5(a) and (c), respectively. They are taken from the 110 pole of $L1_0$ grains, which are seen as dark regions in (a). Distances between spots in (c) correspond to lattice constants of $L1_0$ phase evaluated from the results of XRD. The micrograph (b) is an image of the same view taken from the 110 pole of B2 grains, and (d) is the corresponding SAD pattern. In the diffraction pattern there are many extra spots, and remarks about them will be given later. As is seen in these figures, the stress induced $L1_0$ phase grains appeared as layers of about 100 to 1000nm thickness in the B2 matrix grains. Crystallographic relationships between $L1_0$ and B2 phases were determined by analyses of SAD patterns; $(111)_{B2} // (111)_{L1_0}$, and $[11\bar{2}]_{B2} // [11\bar{2}]_{L1_0}$. Boundaries between two phases are not flat,

Figure 7: Fcc phase grains appeared in a B2 grain. (a) Bright field image of a B2 grain. (b) Dark field image taken from the extra spot pointed by an allow in the SAD pattern in (c).

concentration in the Fe rich region up to 20at%Rh, where B2 ordered phase comes out, and then the transition temperature changes to increase. The existence of a disordered bcc phase is not observed in the alloy with the Rh concentration higher than 30at%. These features suggest that B2 ordering stabilizes bcc type structure in Fe-Rh system. From this viewpoint, the B2-fcc transition may be interpreted as a phenomenon caused by destruction of the order of B2 structure with cold working. In fact, from the result of XRD, the changes of the intensity ratio of $100_{B2}/200_{B2}$ reflection shows a considerable decrease of the degree of order at a rolling rate of 30%, where the fcc reflections begin to develop.

On the other hand, $L1_0$ phase appears at a small rolling rate of 5%. Destruction of order must not play an important role in this case. It is difficult at present to mention the origin of B2- $L1_0$ transition. However, martensitic transformations are usually enhanced by cold working. The $L1_0$ phase may be an enhanced martensite that is not formed with cooling because of a shortage of the driving force.

though it is not able to determine habit plane precisely, they are near $\{100\}$ planes of B2 phase. In Fig.6 is shown internal structures of $L1_0$ grains. $L1_0$ grains are divided into platelet subgrains, each of them has slightly different directions from others. Density of dislocations is extremely high, and transformation twins are not observed. These features show that the B2- $L1_0$ transition is a martensitic transformation accompanying slip deformation.

Until now, we have presented the results on stress induced $L1_0$ phase, and what about fcc? In truth, their diffraction patterns have been already presented. Stress induced fcc phase in an early stage of their growth exist as platelets in B2 matrix grains, and extra spots in Fig.5(d) are from such platelets in the B2 grain (and their double diffraction). Figure 7(a) is a bright field image of a B2 grain in the same specimen. A dark field image of (b) is taken from the spot pointed by an allow in the SAD pattern of (c) corresponding to 220_{fcc} reflection. Platelets of fcc phase sized about a few nanometer are seen in (b). In specimens rolled at a rate of 30%, these platelets commonly appeared in B2 grains, but seldom did in $L1_0$ grains.

4. Discussion

As was reported in previous reports,[4-7] there are two different stress-induced phases obtained from B2 ordered Fe-Rh alloys. It is found from XRD investigation that $L1_0$ phase appears at first in the early stage of cold work, and then it decreases with later appearance of fcc phase. In addition, the $L1_0$ structure seems to be a compromise of B2 and fcc, suggesting that $L1_0$ phase is an intermediate phase of the B2-fcc transition. However, this simple idea is inconsistent with the results of TEM observation. If $L1_0$ is an intermediate phase, the terminal fcc grains must be found in the $L1_0$ phase, but they appear directly from B2 phase, and they are far different from the $L1_0$ grains in their shapes and sizes. Moreover, they are seldom found in the $L1_0$ grains of specimens rolled at a rate of 30%. It means that $L1_0$ phase is harder to change into fcc phase than B2 phase, in reality. Though occurrence of the $L1_0$ -fcc transition with heavy working is obvious from the results of XRD investigation, it must be thought that the B2- $L1_0$ transition and the B2-fcc transition are independent phenomena from each other.

Then, we must think about the origin of each transition.

The B2-fcc transition is a structural change from an ordered phase to a disordered one, and it is obvious that some process of disordering involves in the transition. According to the phase diagram of Fe-Rh system,[1] the bcc-fcc transition temperature decreases with the increase of Rh

5. Conclusions

The stress induced transition of an Fe-50.5at%Rh alloy was investigated with XRD and TEM. The results are summarized as follows:

1. With the progress of cold working, the B2-L1₀ transition occurs at first. The transition to fcc takes place in a later stage, and consequently the L1₀ phase is decreased.
2. The B2-L1₀ transition and the B2-fcc transition are independent each other. The L1₀ phase is not an intermediate phase of the B2-fcc transition.
3. Destruction of the long range order of B2 with the progress of cold working may be a cause of the B2-fcc transition.

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References

- [1]. Massalski T. B., Binary Alloy Phase Diagrams (American Society for Metals, Metal Park, Ohio 44073, 1986) pp.1099-1100.
- [2]. Fallot M., Ann. Phys.(Paris) **10**(1938),291. Fallot M. and Hocart R., Rev. Sci. **77**(1939),498.
- [3]. Schinkel C. J., Hartog R. and Hoshstenbach F. H. A. M., J. Phys. F, Metal Phys. **4**(1974),1412.
- [4]. Lommel J. M. and Kouvel J. S., J. Appl. Phys. **38**(1967),1263.
- [5]. Miyajima H. and Yuasa S., J. Magn. Magn. Mater. **104-107**(1992),2025.
- [6]. Yuasa S. and Miyajama H., Nucl. Instr. and Meth. **B76**(1993),71
- [7]. Miyajima H., Yuasa S. and Otani Y., Jpn J. Appl. Phys. **32**(1993),232
- [8]. Oshima R., Takahashi M., Taniyama A. and Tsunoda Y., "First Order Phase Transition of Fe-Rh Alloys", International Conference on Martensitic Transformations, Monterey 20-24 July 1992, C. M. Wayman and J. Perkins Eds. (Monterey Institute of Advanced Studies, P. O. Box 4427, Carmel, CA 93921, USA, 1993)pp.1211-1216.
- [9]. Takahashi M. and Oshima R., Mater. Trans. JIM **36**(1995),735.