The Influence of Cold Rolling Reduction on r -value and Recrystallization Behavior in Fe–36Ni Alloy

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Recrystallization behavior and grain growth in Fe–36%Ni invar alloy for shadow masks in color cathode ray tubes are studied by using transmission electron microscopy. The r -value increases with increasing (111) surface intensity. In case of light cold-rolling reduction, nuclei orientations of recrystallized structure after annealing are (011) and the nuclei are surrounded by cold rolled matrices of (011) orientation. On the other hand, in case of heavy cold rolling reduction, nuclei orientations of recrystallized structure after annealing are (011) and (100). These two nuclei are surrounded by the same cold rolled matrices of (011) orientation with light cold rolling reduction.

KEY WORDS: TEM; invar; shadow mask; recrystallization; grain growth; annealing; nucleus; r -value; X-ray intensity; texture.

1. Introduction

For a long time, shadow mask for the color cathode ray tubes have been made of Al killed steel sheet. In recent years, it is demanded that the color cathode ray tubes have higher brightness, greater size with good flatness and higher definition. The temperature of the shadow mask rises up to about 313 K when the beam hits the shadow mask. This temperature increase results in the thermal expansion and deteriorates the high definition. Therefore, Fe–36Ni invar alloy that has quite low thermal expansion coefficient is adopted in order to overcome this problem. Besides this property, material of the shadow mask for the color cathode ray tubes must have the good formability and the excellent etching property. Some studies have reported that these properties can be improved with special defined textures that prefer (001), (111), (011) faces on the sheet surface.^{1–4)} In invar alloy, the typical cube orientation texture in fcc metal appears and sheet surface consists mostly of the (001) orientation of recrystallized grain.^{5,6)} Oxidation phenomena during annealing in Fe–36Ni alloy are already investigated.⁷⁾ However, there are only a few papers concerning the recrystallization behavior in Fe–36Ni alloy.

In the present study, the influence of cold rolling reduction and annealing temperature on the r -value related to the formability and the recrystallization behavior in Fe–36Ni invar alloy are investigated.

2. Experimental Procedure

A commercially typical grade of Fe–36Ni alloy for the shadow mask was sampled from cold rolled strips made by conventional mill processing. The chemical composition of the specimen used are listed in **Table 1**. The sheet sample of 0.65 mm in thickness was initially heat-treated at 1023 K for 60 sec. The texture of the samples was found to be (100)[001] and (221)[212] with pole figure method. The grain size number was 8 compared with austenite standard structure. (JIS-0551) The samples were cold-rolled in laboratory to various reduction in thickness from 5 to 80%. In order to simulate the continuous annealing cycle and to observe the recrystallized grains, the sample was treated by two types of the annealing cycles.

(1) The cycle of continuous annealing in laboratory:

Sample was heated at the rate of 10 K/sec, soaked at 873–1 173 K for 1–3 180 sec (53 min).

(2) Salt bath cycle:

Sample was heated at 873–1 173 K, held for 1–3 180 sec (53 min) and then quenched into ice water. This cycle was used for the investigation of TEM observation.

Table 1. The chemical composition of the specimen used in this study. (mass%)

C	Si	Mn	P	S	Al	N	Ni
0.0017	0.056	0.25	0.002	0.0017	0.001	0.003	35.90

Fig. 1. Schematic drawing of the change of hardness with annealing temperature.

The r -value in three directions (parallel, 45 and 90 degree to the rolling direction) was measured by tensile test. The measurement position of the X-ray reflection intensity of the plane parallel to the surface was the center of the thickness and X-ray was irradiated from the normal direction. To select the specimen for TEM observations of recrystallized structure, Vickers hardness was measured in the specimen annealed under various conditions. The schematic drawing of hardness changes is shown in Fig. 1, which are divided into three stages, *i.e.* (1) early stage, (2) finishing stage, (3) full annealing stage. Disk specimens of 3 mm in diameter corresponding to each stage were electropolished using twin jet method. The electrolyte consisted of chromic-acid, acetic-acid and pure water. Selected area diffraction experiment and TEM observations were carried out in a JEOL-2010 microscope operated at 200 kV.

3. Results and Discussion

3.1. Effect of Grain Orientation on the r -value

Figure 2 shows the effect of cold rolling reduction on the r -value. The samples were cold-rolled with various reduction rates of 5 to 78% and subsequently annealed at 1073 and 1173 K for 100 sec. In case of the annealing temperature at 1073 K, the r -value had max. value on the 33% reduction and decreased with increasing reduction. In case of the annealing temperature at 1173 K, the r -value reached maximum value at the 20% reduction and also decreased with higher reduction.

The influence of cold rolling reduction on X-ray intensity are shown in Fig. 3. In as rolled samples, (220) intensity increases as the reduction is increased. Owing to this, (111) and (200) intensities decreases. On the other hand, in case of the annealing at 1073 K for 100 sec, the behavior of the (111) intensity is similar to that of as rolled sample in the reduction range lower than 15% reduction. That is, (111) intensity decreases with reduction less than 15% in the same manner as the same sample as rolled and then increases with reduction higher than 15%. The maximum value of (111) intensity is obtained at 33% reduction. Finally, it decreases gradually with reduction higher than 33%. It is considered that the recrystallization do not happen in the reduction range lower than 15%, since the behavior of the (111) intensity is similar to that of as rolled samples.

(200) intensity is quickly increasing from 68% reduction to 78% reduction. (220) intensity reaches the

Fig. 2. Effect of cold rolling reduction and annealing temperature (period: 100 sec) on r -value.

Fig. 3. Effect of cold rolling reduction and annealing temperature (period: 100 sec) on X-ray reflection intensities.

maximum value at the 20% reduction and in the range of upper 20% reduction and then decreases with increasing cold rolling reduction. In case of annealing at 1173 K for 100 sec, (111) intensity starts increasing from 15% reduction and reaches the maximum value with 20% reduction. (111) intensity decreased slowly with increasing reduction higher than 20%. (200) intensity starts increasing from 15% reduction and (220) intensity get the max. value on 33% reduction. The behavior of annealing at 1073 K is similar to that of annealed at 1173 K.

The relationships between r -value and ND plain intensities in deep drawing quality of low carbon steels have been investigated in some reports. r -value can be improved with increasing (111) intensity, and (200) and (220) intensities deteriorates the r -value.^{8,9)} In the present

study, from Figs. 2 and 3, it is found that the r -value is not related to the (200), (220) intensities, but strongly influenced by (111) intensity. The reason why the r -value decreased in the range of cold rolling reduction 10 to 15% is supposed that the recrystallization should not take place in this conditions. (111) intensity is too weak to have the effect on the r -value. When annealing temperature is enough high for the recrystallization, r -value increases as the (111) intensity increases. In heavy cold rolling reduction, it is considered that the r -value decreased because (200) intensity increased rapidly. On the contrary, (111) intensity decreased. **Figure 4** shows the relationships between mean r -value and (111), (200) and (220) intensities respectively. These two factors, mean r -value and (111) intensity shows strongly correlation. But, (200) and (220) intensities are not correlated with mean r -value. The relation between mean r -value and three intensities is expressed by the following equation, which is defined from regression analysis.

$$r = 0.232I(111) - 0.004I(200) + 0.011I(220) + 0.821$$

here r : mean r -value, $I(111)$: (111) intensity, $I(200)$: (200) intensity, $I(220)$: (220) intensity.

Fig. 4. Relationship between r -value and (111) intensity.

The regression coefficient is 0.81. From Fig. 4, in these intensities, (111) intensity has a strong relationship with mean r -value. The relation between mean r -value and (111) intensity is expressed by the following equation

$$r = 0.29I(111) + 0.77$$

here r : mean r -value, $I(111)$: (111) intensity.

The regression coefficient is 0.74. On the other hand, the regression coefficient between mean r -value and (200) intensity was 0.50 and that between mean r -value and (220) intensity was 0.007.

3.2. Recrystallization Behavior

As mentioned above, it is apparent r -value increases with increasing (111) intensity. With heavy cold rolling for fcc metal, the representative cube texture develops. In the present Fe-36N alloy, (220) intensity is also pretty high with heavy cold rolling.

Therefore, in order to investigate the difference of recrystallization behavior between heavy and light cold rolling reduction, the recrystallized nuclei and grains are observed by TEM.

3.2.1. Early Stage of Recrystallization

It is defined that the recrystallized nuclei are the one or two grains appeared from the cold rolling matrix. The 33% reduction for the light rolling reduction and the 78% reduction for the heavy reduction are selected. The samples of 33% reduction are annealed at 1073 K and that of 78% reduction are annealed at 923 K, respectively. The ratio of the recrystallization in the early stage is approximately 6% in both 33 and 78% cold rolling reductions.

(1) Heavy (78%) Cold Rolling Reduction

Figure 5(a) shows the typical TEM micrographs of the recrystallized nuclei with 78% reduction. Face orientation of recrystallized nucleus are (001) orientation and

Fig. 5. TEM micrograph of the annealed nucleus.
(a) 78% cold rolling reduction and annealed at 923 K
(b) 33% cold rolling reduction and annealed at 1073 K

it is surrounded by (011), (112) and (123) cold rolled matrices. From other foil samples, (011), (013) and (112) face orientation of recrystallized nuclei are observed and especially (011) face orientation of nuclei is 58% in all observed nuclei with 78% reduction as described below in Fig. 7.

(2) Light (33%) Cold Rolling Reduction

With 33% reduction in Fig. 5(b), face orientation of the recrystallized nuclei is (011) orientation and it is surrounded by (122), (011), (013), (112) and (111) cold rolled matrices. From other foil samples, (011), (013) and (112) face orientation of recrystallized nuclei are observed and (111) nucleus is seldom observed. In the reference, (112) orientation nuclei and (112) cold rolled matrix are observed as the main orientation in Fe-30Ni alloy,¹⁰⁾ but in the present study, they are not regarded as the main orientation. Although it is expected that the (111) face orientation of recrystallized nucleus are observed, there are only 2% in the samples annealed at 1073 K with 33% reduction as described below. On the other hand, many regions consists of some recrystallized grains in the cold rolled matrix are observed in the identical samples with 33% reduction in early stage. In these regions, (111) face orientation of recrystallized nuclei can be seen. It is confirmed by the existence of (111) orientation of recrystallized grains. It is considered that the (111) face orientation of recrystallized nuclei is not

formed but grains of (111) orientation of recrystallized nuclei should be come up in the grain growth process. In order to investigate the influence of the annealing temperature on the orientation of recrystallized nuclei, the sample of 33% reduction was annealed at 923 K for 1800 sec (30 min). TEM micrographs of recrystallized nuclei is shown in Fig. 6. (011) face orientation of recrystallized nucleus are observed and it is surrounded by (011) and (114) cold rolled matrices. In other foil samples, (011), (112) and (013) orientation of nuclei are also observed and are surrounded by (011), (001) and (112)

Fig. 6. TEM micrograph of the annealed nucleus. Both (a) and (b) are cold rolled with 33% reduction and annealed at 923 K.

Fig. 7. Stereographic unit triangles showing the orientation distribution in the early stage.

orientation of recrystallized nuclei.

Figure 7 shows the stereographic unit triangles summarizing the face orientation of recrystallized nuclei and grain in early stage of the recrystallization.

In case of 78% reduction with annealed at 923 K for 100 sec, (011) orientation of recrystallized nuclei and typical (001) orientation of recrystallized nuclei as seen in fcc metal are main orientation of recrystallized nuclei. These recrystallized nuclei are surrounded by (011), (112) cold rolled matrices. In generally, (001)[100] cube orientation of recrystallized structure appears with heavy cold rolling in fcc metal. Judging from the X-ray reflection intensity, the (011) face orientation of grains is pretty high. (011) orientation grains were also formed in early stage of recrystallization. (011) orientation nuclei are surrounded by (011) cold rolled matrices. In Fe-36Ni alloy, it is found that the typical cube orientation texture are formed by both rolling reductions.

In the light cold rolling reduction with annealed at 1073 K for 15 sec, many (011) orientation of recrystallized nuclei are observed as described below. (011) orientation of recrystallized nuclei is surrounded by (012), (114) orientation cold rolled matrices. (013) orientation of recrystallized nuclei is surrounded by (114), (125) orientation cold rolled matrices. (112) orientation of recrystallized nuclei is surrounded by (011), (114), (123) orientation cold rolled matrices. (111) orientation of recrystallized nucleus is observed only 2%. It is considered that the reason why (111) orientation of recrystallized nucleus is seldom observed is due to the relatively high annealing temperature at 1073 K. So it is tested using the low annealing temperature at 923 K.

Figure 7 shows the results of orientation of recrystallized nuclei and information of the surrounded cold rolled matrices. In the light cold rolling reduction with annealed at 923 K for 1800 sec (30 min), TEM micrographs are shown in Fig. 6. (011) orientation of nucleus is observed and is surrounded by (114), (011) cold rolled matrices. In other foil samples, (011) orientation nuclei is observed 76%, indicated in Fig. 7, and was surrounded by (011), (001) cold rolled matrices. It is thought that the (001) cold rolled matrix was remained because of light cold rolling. Although it is expected that the (111) orientation nucleus was formed with lower annealing temperature at 923 K, (111) orientation of recrystallized nucleus are not observed. On the contrary, (011) orientation of recrystallized nuclei increases compared with using high annealing temperature.

3.2.2. Finishing Stage of Recrystallization

(1) Heavy Cold Rolling Reduction

Figure 8 shows the TEM micrograph and schematic illustration of the 78% cold rolling reduction with annealed at 923 K for 3180 sec (53 min). (011), (001), (112), (123) and (013) orientation of recrystallized grains are observed. From the results of other foil samples, there are many (011) and (001) orientation of recrystallized grains as described below in Fig. 10. From the TEM image of Fig. 8, (011) orientation of recrystallized grains were 7 grains in all observed 11 grains.

Fig. 8. TEM micrograph and schematic drawing of the annealed grain with cold rolled 78% reduction annealed at 923 K for 3180 sec (53 min).

(a) TEM micrograph, (b) Schematic drawing

Fig. 9. Schematic drawing of the annealed grain with cold rolled 33% reduction annealed at 1073 K for 100 sec.

(2) Light Cold Rolling Reduction

Figure 9 shows the schematic diagram of the specimen rolled with 33% light reduction and annealed at 1073 K for 100 sec. In this figure, (011), (111), (001), (112), (114) and (123) orientation of recrystallized grains are observed. However, (111) orientation of recrystallized grain is not observed in the specimen rolled with 78% reduction. As previously mentioned, X-ray reflection intensity of (111) could be high with light cold rolling reduction. In addition, increasing of the (111) orientation of recrystallized grains with using the light cold rolling

Fig. 10. Stereographic unit triangles showing the orientation distribution in the finishing stage and full annealing stage.

Fig. 11. Stereographic unit triangle showing the orientation distribution in the finishing stage in order to obtain intensified (111) face intensity.

reduction is confirmed by the TEM observation. Therefore, higher (111) intensity can be obtained in the specimen which is repeatedly cold-rolled with light cold rolling reduction and annealing. Figure 11 shows the results of the repeated cold rolling and annealing. (111) orientation of recrystallized grains are observed with higher ratio than the single cold rolling and annealing process as mentioned above.

The recrystallization phenomena is summarized in Fig. 10. In case of 33% reduction, (011) orientation of recrystallized gains is 29% and (112) is 24%, respectively. In comparison with heavy cold rolling condition, orientations of grains of the sample with light cold rolling reduction is distributed widely. (111) orientation is 11% in the full annealing stage and that in the specimen of repeated cold rolling and annealing is observed 22%. In case of 78% reduction, (011) and (001) orientation of recrystallized grains accounts for 70% of all observed grains in finishing stage.

3.2.3. Full Annealing Stage of Recrystallization

As already indicated in Fig. 10, the microstructure in the full annealing stage is observed by TEM. Experimental condition was as follows.

Both Light (33%) and heavy (78%) cold rolling condition: annealed at 1173 K for 3810 sec (53 min.)

(1) Heavy Cold Rolling Reduction

(011) and (001) orientation of recrystallized grains accounts for 90% of all observed grains in full-annealing stage as above mentioned in Fig. 10. (001) orientation of recrystallized grain accounts for 66% of all observed grains in this stage even a sole orientation. From the point of view of both finishing and full annealing stages, (001) orientation of recrystallized grains is increased from 25 to 66% with decreasing (011) orientation of recrystallized grain. The result from the measurement of the X-ray intensities is in good agreement with the observation result by TEM, because the (011) intensity rapidly decreases with reduction higher than 68%. It is found that the typical (001) face orientation was strongly appeared when the heavy cold rolling reduction is performed in Fe-36Ni invar alloy. (111) orientation of recrystallized grain with 78% cold-rolling reduction was not observed in this full annealing stage. Therefore, (111) orientation of recrystallized nuclei and grains is not formed in the process of recrystallization in Fe-36Ni alloy with heavy cold rolling reduction.

(2) Light Cold Rolling Reduction

In light cold rolling reduction, (011) orientation accounts for 40% of all grains in full annealing stage (see Fig. 10). The distribution of the orientation in full annealing stage is similar to that in finishing stage. The orientation of the recrystallized grains are distributed widely compared with heavy cold rolling reduction. (111) orientation of recrystallized grain is also observed in the ratio of 9%.

3.2.4. Effect of Cold Rolling Reduction on the Recrystallization Behavior

(1) Heavy Cold Rolling Reduction

In the early stage of recrystallization with 78% heavy cold rolling reduction, (011) and (001) orientations of recrystallized nuclei are observed and mainly formed from (011) cold rolled matrix. Both orientations are surrounded by (011) cold rolled matrix. (001) orientation of recrystallized grains increases with grain growth during in the finishing stage and full annealing stage. In the full annealing stage, (001) and (011) orientation of recrystallized grains accounts for 66 and 24%, respectively. (011) orientation of recrystallized grain is decreased. Therefore, finally, (001) orientation of recrystallized grain as it is called cube texture should be appeared with more prolonged annealing than the annealing condition used in full annealing stage.

(2) Light Cold Rolling Reduction

At first, in the early stage of recrystallization with light cold rolling reduction, (011) orientation of recrystallized nucleus are mainly formed from (011) cold rolled matrix. The orientations of recrystallized nuclei and grains are widely distributed compared with heavy cold rolling reduction. The tendency is not changed even the recrystallization is reached the finishing stage and full-annealing stage. It is considered that the recrystalliza-

tion was controlled by the process that has the mechanism which does not change the orientation from the matrices to recrystallized nuclei. It would be caused by light cold rolling reduction. Second, (111) orientation of recrystallized nucleus is hardly observed. But, (111) recrystallized grains are observed in the identical samples that are used in the observation of the early stage with 33% reduction. That region is in the groups that consisted of several or several decades of recrystallized grains. Both the finishing and full annealing stages, (111) orientation is 11 and 9%, respectively. Therefore, (111) orientation of recrystallized nucleus is not formed but formed during the grain growth of recrystallization.

4. Conclusion

The influence of cold rolling reduction on r -value and recrystallization behavior in Fe-36Ni alloy is investigated. The results of the present study are summarized as follows.

(1) After annealing, (111) intensity increases with light cold rolling and (200) intensity increases with heavy cold rolling.

(2) (111) intensity affects r -value and r -value is increased with increasing (111) intensity.

(3) r -value has a maximum when light cold rolling and relatively high temperature annealing are applied. (111) intensity increases by applying this process repeatedly.

(4) In the combination of light cold rolling and annealing, almost all recrystallized nuclei have (011) orientation and they grow in the (011) cold rolling matrices.

(5) In the heavy cold rolling and annealing, (011) orientation of recrystallized nuclei and (001) orientation of recrystallized grains are observed.

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