Cold Rolling and Annealing Textures in 3.25% Al-Fe Alloy Sheets with or without Sulfur Addition*

By Hitoshi Nakaé** and Noriaki Goshi***

The recrystallization and grain growth in cold rolled 3.25% Al-Fe alloy sheets were studied with the specimens of three different sulfur contents.

On primary recrystallization, the textures were chiefly composed of the components found in as-rolled textures being those of (112)[1 $\bar{1}$ 0], (111)[11 $\bar{2}$] and (100)[011], and (110)[001], and (100)[001] components were also found as minor ones, in which the (110)[001] component showed a tendency to become strong with increasing sulfur content.

During grain growth, the (100)[001] component seemed to increase preferentially consuming the other oriented matrix components and the ratio of the (100)[001] to the (110)[001] component increased and the value tended to approach a definite value.

It was also made sure that the textures as-rolled and of primary recrystallization of Al-Fe alloys were the same as those of cold rolled 3.25% Si-Fe alloy as well as iron sheets, whereas the main components shifted towards the major (100)[001] + (110)[001] components during grain growth in contrast to those of Si-Fe alloy whose texture was composed mainly of the (110)[001] component.

Heating rate had an influence on the development of texture and the presence of a liquid phase in grain boundaries was assumed in the discussion of grain growth mechanism in the alloy with the sulfur addition.

(Received December 27, 1973)

I. Introduction

The recrystallization texture of a cold-rolled 3.25% Al-Fe alloy sheets has been known to take a (100)[001] orientation as the main component (cube texture⁽¹⁾⁽²⁾) in contrast to that of 3.25%Si-Fe alloy sheets. However, it shows similar physical properties with regard to electrical resistivity, magnetic permeability and mechanical tensile behavior.

In the textures of Si-Fe alloy, a cube texture is often revealed under certain conditions, e.g. by recrystallization at an appropriate reduction⁽³⁾, by grain growth in an high pure alloy⁽⁴⁾ and by annealing at temperatures exceeding 1200°C^{(5)~(7)} or by annealing a very thin sheet in a controlled atmosphere⁽³⁾.

In the grain growth of Si-Fe alloy sheets, the main component of the texture is known to be strengthened by a small addition of other elements. The added elements are titanium, niobium, sulfur, manganese and manganese sulfide, and the effects of sulfur and manganese have been reported by May, Turnbull, Ainslie et al. and one of the present authors^{(8)~(17)}.

It would, therefore, be worthwhile to discuss the effects of sulfur on the grain growth of cold-rolled Al-Fe alloy sheets with special reference to the results obtained so far in a cold-rolled Si-Fe alloy and elucidate the underlying mechanism which gives rise to the difference in growth behavior between the two alloys.

Table 1 Chemical composition of the Fe-Al alloy sheet specimens (wt%).

Specimens	Al	C	Si	S	P	Bal.
No. 1	3.17	0.007	0.01	0.004	0.002	Fe
No. 2	3.17	0.009	0.01	0.060	0.002	Fe
No. 3	3.17	0.030	0.04	0.148	0.002	Fe

II. Experimental

The specimens were prepared by melting in a vacuum furnace by adding different amounts of sulfur to a 3.17%Al-Fe base alloy. After hot working, they were cold rolled to a final thickness of 0.35 mm with the reduction of about 75%. The chemical compositions are listed in Table 1, in which the contents of sulfur are 0.004, 0.060 and 0.148% in specimens Nos. 1, 2 and 3 respectively. No sulfur was added in specimen No. 1 and the high content of carbon in specimen No. 3 was due to the carbon contained in the added material sulfur which would give no substantial effect on the results.

The specimens were annealed at 50°C intervals from 600°C or 100°C intervals from 700°C up to 1200°C by isothermal annealing for 1 hr. Alternatively, 10°C or 20°C intervals were employed at the specified temperatures. An optical microscope, a goniomicroscope⁽¹⁸⁾, an X-ray diffractometer and a torque magnetometer were used for the observation of structures, and determination of the crystal orientation, and textural analysis, respectively.

The basis for the analysis of the texture of a polycrystalline specimen by the torque magnetometer will be given briefly in the following⁽¹⁹⁾. The magnetic torque of the polycrystalline specimen is a statistical average over the basic curves for crystals of several orientations, in which the influential orientations on

* The main part of this paper was originally published in Japanese in J. Japan Inst. Metals, 33 (1969), 553.

** Department of Precision Engineering, Faculty of Engineering, Hokkaido University, Sapporo 060, Japan.

*** Graduate School, Hokkaido University, Sapporo. Present address: Hitachi Kinzoku Manufacturing Co., Kawagoé, Japan.

Fig. 1 Magnetic torque curves for single crystals of typical orientations (a), and those of specimens as-rolled and annealed at 800°C for 1 hr (b), and maximum torque L_{m1} and L_{m2} .

the curves are a few components such as (110) [001], (100) [001], and (100)[011].

For the (100)[001] crystal, the magnetic torque L is given by

$$L = -(K_1/2) \sin 4\alpha,$$

for the (100)[011] crystal which is one of the components of the deformation texture and an orientation obtained by a rotation of 45° from the above crystal,

$$L = (K_1/2) \sin 4\alpha,$$

and for the (110)[001] crystal,

$$L = -(K_1/4) \sin 2\alpha - (3K_1/8) \sin 4\alpha,$$

where K_1 is the first anisotropic constant, α the direction cosine of [001] direction with respect to the X axis of the three-dimensional coordinate. In this case, other direction cosines with respect to the Y and Z axes disappear. The second anisotropic constant K_2 was neglected because of its meager contribution to the torque curve. In this paper, only the development of the (100)[011] and (110)[001] components have been discussed. Other components which might have a minor effect are neglected.

The torque curves of (110)[001] and (100)[001] orientations are approximately in phase so that the maximum torque values are associated with the degree of alignment of the (110)[001]+(100)[001] components. The (100)[011] component is anti-phase to the latter two components; hence its existence in the texture diminishes the maximum torque values distinctly. As for the quantitative proportionality between the (110)[001] and the (100)[001] components in the texture, the ratio of the maximum values of the

first to the second may be estimated, since they are 0.43 and unity with (110)[001] and (100)[001] orientations respectively. The magnetic torque curves taken for the specimens over the entire range of recrystallization so far showed the characteristic shape of the (110)[001]+(100)[001] components and the analysis of which will be made.

III. Results

Using an X-ray diffractometer, the textures of the three specimens with various sulfur contents were determined after cold rolling, primary recrystallization and grain growth. A qualitative analysis of the shift rate of the texture during grain growth was made using the magnetic torque curves and the X-ray diffractographs.

1. Textures as-rolled

The pole figures are shown in Figs. 2(a), (b) and (c). The textures are shown to consist of the three major components of (112)[110], (111)[112] and (100)[011].

2. Primary recrystallization textures

The primary recrystallization textures of the three specimens are shown in Figs. 3(a), (b) and (c). They were annealed at 700°C for 1 hr, by which they seemed to be at a step generally expressed as primary recrystallization. The components constituting the textures are the same as those of the cold rolled textures with a rather increased spread of (112)[110], (111)[112], (110)[001], (100)[001] and a fairly large part of random components, although the (112)[110], (111)[112] and (110)[001] components become sharper by the addition of sulfur. In the optical microscopic observation the majority of the (110)[001] and (100)[001] components were the recrystallized grains, whereas the others were those at a recovery stage.

The cold rolled and primarily recrystallized textures of 3.25%Al-Fe alloy may be considered to be substantially similar to those observed in the cold rolled 3.25% Si-Fe alloy under the same effect of sulfur.

In a 3.25%Si-Fe alloy sheet, the recrystallized grains of the same orientation appeared as a group forming a colony or a flora, and among them the grains of (110)[001] and (100)[001] orientations existed near the surface of the sheet with a small orientation spread, whereas those of (111)[112] and (112)[110] orientations existed in the inside of the specimen with a rather large orientation spread⁽¹⁷⁾. Thus, the formers are known to grow dominantly against the latters at the stage of grain growth. This behavior may also presumably occur in the recrystallization of the cold rolled Al-Fe alloy sheet.

3. Textures during and after grain growth

(1) Pole figure

The pole figures of (110)- and (100)-poles on the specimens annealed at 1200°C for 1 hr are shown in Figs. 4(a), (b) and (c), and Figs. 5(a), (b) and (c) respectively. From these figures, it is concluded that

Fig. 2 (110)-pole figures of 3.17% Al-Fe alloy sheet as-rolled. Δ , $\circ$ and $\square$ indicate (111)[112], (112)[110] and (100)[011]-poles respectively.

the growth of both the (110)[001] and (100)[001] components occurs at the expense of the other residual components, in contrast to the 3.25%Si-Fe alloy sheet whose growth component is simply a (110)[001] orientation.

In specimen No. 1, the texture is composed of two components of (110)[001] and (110)[001] with a weak (112)[110], (111)[112] and dispersed (100)[011]. In specimen No. 2, the main components are the same as in No. 1, but the other components are hardly observable. In specimen No. 3, the main components are also the same as in the above two specimens, but the (111)[112] and (100)[011] components are seen at a very small degree. In these three specimens, the (110)[011] and (100)[001] components which form the main part of the texture are seen with a slight dispersion of orientation around the common [001] axis.

(2) Texture analysis by magnetic torque curves

The magnetic torque curves were taken on the specimens annealed at temperatures between 600 and 1200°C and the tendency of the textural shift during

Fig. 3 (110)-pole figures of 3.17% Al-Fe alloy sheet annealed at 700°C for 1 hr in hydrogen atmosphere.

grain growth was observed quantitatively.

In the primary recrystallization, as seen in Figs. 6 (a) and (b), the maximum torque values decrease with the increased sulfur addition, while the torque ratios are large and almost the same between specimen Nos. 1 and 2, and small in No. 3. They can be analysed, together with the pole figures in Fig. 4, as having (110)[001], (100)[001], (111)[112] and the (100)[011] components which exist commonly in these three specimens and the former two components occupy a large portion of the specimens in which the (110)[001] is the most dominant.

To elucidate the tendency of the quantitative proportionality for the (110)[001] and (100)[001] components in the textures during the growth, the torque ratio vs temperature curves in Fig. 6(b) have to be considered. An increase of the torque ratio would mean that the (100)[001] component was dominant over the (110)[001] component during grain growth. Here,

Fig. 4 (110)-pole figures of 3.17% Al-Fe alloy sheet annealed at 1200°C for 1 hr in hydrogen atmosphere.

the growth of the two major components can be achieved by absorbing the other components such as $(111)[11\bar{2}]$, $(112)[1\bar{1}0]$, $(100)[011]$ and others existing in the primary recrystallization texture. These results may be characteristic of the Al-Fe alloy and somewhat different from those of Si-Fe alloy whose dominant growth component has a $(110)[001]$ orientation⁽¹⁵⁾.

At the ascending temperature, the maximum torque values show at first a direct increase in the specimen No. 1 of without sulfur addition, it remain unvaried up to 800 and 1000°C in specimen No. 2 with a moderate level of sulfur addition and in specimen No. 3 with the highest level of sulfur addition. A retardation of growth beginning at 900°C is observed in specimen No. 1, whereas no retardation is seen in specimen Nos. 2 and 3.

It is to be noticed that the torque maxima and

Fig. 5 (100)-pole figures of 3.17% Al-Fe alloy sheet annealed at 1200°C for 1 hr in hydrogen atmosphere.

torque ratios, i.e. the concentrations of $(110)[001]$ + $(100)[001]$ components and their proportionalities in the textures approach generally the similar values in both alloys with or without sulfur addition regardless of their intermediate values with temperatures (Fig. 7).

Until the growth stage of annealing at 1200°C, the $(112)[1\bar{1}0]$ and $(111)[11\bar{2}]$ components which constituted the main part of the as-rolled textures remain slightly in specimen No. 1 and to a considerable extent in specimen No. 3; the presence of these components is scarce in specimen No. 2.

IV. Discussion

The experimental results obtained for the influence

Fig. 6 Maximum torque value and peak value ratio vs isochronal annealing temperature for 1 hr with heating rate of about 6°C/min. ○, □ and × indicate plots of specimens No. 1, No. 2 and No. 3 respectively.

Fig. 7 Peak value ratio vs maximum torque. ○, □ and × indicate plots of specimens No. 1, No. 2 and No. 3 respectively.

of sulfur on the recrystallization textures of the cold-rolled 3.17%Al-Fe alloy may be discussed as follows.

The cold-rolled textures consisted of $(112)[\bar{1}\bar{1}0]$, $(111)[11\bar{2}]$ and $(100)[011]$ components regardless of sulfur content as in the case of the Si-Fe alloy and their intensities increased with an increase in sulfur content.

The primary recrystallization textures were composed of $(112)[\bar{1}\bar{1}0]$, $(111)[11\bar{2}]$ and $(100)[011]$ with a slightly wider spread than those of the as-rolled textures. The $(110)[001]$ and $(100)[001]$ components were observed by an optical microscope as minor but distinctly recrystallized ones.

The textures of as-rolled and of primary recrystallization of Al-Fe alloy may, therefore, be said to be the same as those of Si-Fe alloy as well as those of unalloyed iron whose as-rolled and primary recrystallization textures consisted of $(100)[011] + (112)[\bar{1}\bar{1}0]$

Fig. 8 Maximum torque value and peak ratio vs annealing temperature at the heating rate of 2°C/min. ○ and × indicate specimens No. 1 and No. 3 respectively.

and $(111)[11\bar{2}] + (100)[011] + (112)[\bar{1}\bar{1}0]^{(20)(21)}$.

The grain growth of the alloys was characterized by the growth of the recrystallized components of $(110)[001]$ and $(100)[001]$ and the decay of the components of $(111)[11\bar{2}]$, $(112)[\bar{1}\bar{1}0]$, $(100)[011]$ etc. which were regarded as residual components built in cold rolling. In all the specimens, the $(110)[001] + (100)[001]$ components developed with the increasing growth rate of the $(100)[001]$ component rather than the $(110)[001]$ component. The intensities of the $(110)[001] + (100)[001]$ components and the proportionalities of these two components attained nearly the same values in the alloys with or without sulfur addition at the same heating rate. This is in contrast to the Si-Fe alloy in which the growth enhanced the $(110)[001]$ component. It seems that in the Al-Fe alloy the sulfur addition caused the growth stage to shift toward the higher temperature side rather than to modify the growth habit. In the temperature range of growth, the region of exceeded sulfur concentration originated possibly by grain boundary migration, could liquefy at the point corresponding to the eutectic temperature of the Fe-S phase⁽¹²⁾⁽¹⁷⁾. Then, the mode of growth may come to be controlled in such a way as specified "in the presence of a liquid phase" whose thought was first introduced by Kingery et al. for the grain growth of polycrystalline materials in a solid state^{(22)~(24)}. In this case, the process may proceed in the situation which involves both the tendency to deviate the sulfur concentration from the nominal one and the thermal fluctuations to bring it back again to the nominal one. Therefore, the factors such as the kind and concentration of the added elements and heating rate should affect the growth behavior of alloys. In Figs. 8(a) and (b) of growth curves expressed by magnetic torque at the heating rate of 2°C per minute, the torque maximum attained a very high value in the alloy with sulfur addition, while the torque ratio was almost the same as that of the alloy without sulfur addition. This means that a very high

alignment of (110)[001]+(100)[001] components was achieved in the alloy with sulfur addition by the same constitutional proportionality of the components as in the alloy without sulfur addition. This also means that the other components existed in the primary recrystallization texture were nearly absorbed by the above two components.

It is of interest to note that the intensity of the cube component increases by partially replacing silicon by aluminum in Si-Fe alloy retaining the total amount of Si+Al constant^{(25)~(27)}.

V. Conclusion

The results on the texture formation of cold rolled 3.17%Al-Fe alloy with three different contents of sulfur can be summarized as follows.

(1) The cold rolled textures were composed of the same orientations regardless of sulfur content and were the same as those of 3.25%Si-Fe alloy as well as unalloyed iron.

(2) The primary recrystallization textures consisted of the main components existed in the as-rolled textures with a slight spread and of the distinctly recrystallized components of (110)[001] and (100)[001].

(3) On grain growth, the above two recrystallized components developed with the increased growth rate of the (100)[001] component with temperature and attained a similar growth level of texture showing the same torque maxima and torque ratios in the alloys with or without sulfur addition at the same heating rate.

(4) The high concentration of the sulfur in the grain boundaries was expected and thereby the mode of grain growth in the presence of a liquid phase occur.

REFERENCES

- (1) R. M. Bozorth, H. J. Williams and R. J. Morris: *Phys. Rev.*, **58** (1950), 203.
- (2) R. M. Bozorth: *Ferromagnetism*, Van Nostrand., New York, (1953), p. 219.
- (3) K. Detert: *Acta Met.*, **7** (1959), 589.
- (4) C. G. Dunn and J. L. Walter: *Trans. Met. Soc. AIME*, **218** (1960), 448.
- (5) G. Wiener: *J. Appl. Phys.*, **35** (1964), 856.
- (6) F. Assmuss, K. Detert and G. Ibe: *Z. Metallk.*, **48** (1967), 344.
- (7) G. Wiener, P. A. Albert, R. H. Trapp and M. F. Littmann: *ibid.*, **29** (1958), 366.
- (8) H. Fiedler: *Trans. Met. Soc. AIME*, **221** (1961), 1201.
- (9) H. Fiedler: *ibid.*, **227** (1963), 776.
- (10) H. Fiedler: *J. Appl. Phys.*, **29** (1958), 361.
- (11) E. W. Walter and J. Howard: *J. Iron Steel Inst. (London)*, **194** (1960), 96.
- (12) N. G. Ainslie and U. Seybolt: *ibid.*, **194** (1960), 341.
- (13) J. May and D. Turnbull: *Trans. Met. Soc. AIME*, **212** (1958), 769.
- (14) J. May and D. Turnbull: *J. Appl. Phys.*, **30** (1959), 210.
- (15) B. R. Banerjee, R. E. Lenhart and W. H. Robinson: *J. Appl. Phys.*, **33** (1962), 1224.
- (16) J. W. Flower, Jr. and S. P. Karas: *J. Appl. Phys.*, **38** (1967), 1085.
- (17) H. Nakaé and K. Tagashira: *Trans. JIM*, **14** (1973), 15.
- (18) T. Taoka, E. Furubayashi and S. Takeuchi: *Jap. J. Appl. Phys.*, **4** (1965), 120.
- (19) For example: F. Brailsford: *Magnetic Materials*, Methuen Publ. Co., London, (1960), p. 65.
- (20) R. G. Aspdon, T. A. Bergers and R. S. Mateer: *Trans. ASM*, **25** (1937), 1084.
- (21) G. Kurjumov and G. Sachs: *Z. Phys.*, **62** (1930), 592.
- (22) W. D. Kingery: *J. Appl. Phys.*, **30** (1959), 301.
- (23) W. D. Kingery and M. D. Narasimhan: *ibid.*, **30** (1959), 307.
- (24) W. D. Kingery, E. Niki and M. D. Narasimhan: *J. Amer. Ceram. Soc.*, **44** (1961), 29.
- (25) H. Meyer and H. Schlüter: *Stahl u. Eisen*, **73** (1953), 1706.
- (26) F. Loos and H. Schrader: *ibid.*, **73** (1953), 1715.
- (27) I. Gokyu, H. Abe, T. Hashida and S. Takahashi: *J. Japan Inst. Metals*, **25** (1961), 712 (in Japanese).