The Grain Growth after Light Cold Rolling in Grain-Oriented Silicon-Iron Sheets*

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The grain growth after light rolling below 10 % in grain-oriented silicon-iron sheets containing 3.50 and 1.43 % Si was studied optically and magnetically in order to obtain fundamental data for the establishment of a strain-annealing procedure as a modification of the Goss process. It has been found that, in initially fine-grained sheet specimens, the grain growth increases more prominently at two reduction rates, namely, at 0.5 to 2 % and 6 to 7 %. Their resulting preferred orientations are different; in the former, a marked development of the Goss texture occurred, possibly involving no primary recrystallization, while in the latter recrystallization from the Goss texture also occurred.

The recrystallization from the Goss texture occurred also on initially coarse-grained specimens. It has been found that the optimum point for producing silicon-iron sheets with high permeability and low hysteresis loss is at the first grain growth maximum of 2 % reduction for initially fine-grained sheet specimens.

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I. Introduction

Goss⁽¹⁾ reported first in 1935 a method of producing grain-oriented silicon-iron sheets with grains of a (110) plane parallel to the rolling plane and a [001] direction parallel to the rolling direction, this grain texture being now commonly called the Goss texture. Goss's procedure is to cold-roll purified silicon-iron sheets containing 3 to 3.5 % Si to 30 to 40 % of their original thickness, repeating the same cold rolling again after an intermediate annealing at 850 to 900°C, and finally to anneal them at about 1100°C in pure dry hydrogen.

In 1947, Jackson⁽²⁾ and, independently, the present author⁽³⁾ presented a modification of the Goss procedure, in which silicon-iron sheet already cold-rolled to 30 to 40 % of their original thickness and then annealed at 850 to 900°C, namely, subjected to the first stage of the Goss procedure, was again cold-rolled by a small percentage and annealed at 850°C in pure hydrogen. The last stage of our procedure is similar to the so-called strain-anneal technique for growing single crystals and so the author's procedure is, so to speak, the first stage of the Goss procedure plus the strain-anneal procedure. Our procedure produces a well developed, Goss texture with a large grain size, and it is now used in many steel mills. But little is known as to the behavior of crystal grains during the strain-anneal stage of our procedure, which is the subject of the present study.

II. Specimens and Experimental Procedure

The specimens used for the present study are 3.50 and 1.43 % silicon-irons, of which the chemical compositions are shown in Table 1. The starting materials are 1.6 mm thick sheets as produced by hot rolling from

ingots. These sheets were annealed at 800°C, cold rolled to 0.35 mm in thickness, and then annealed at 900°C in pure dry hydrogen, namely, subjected approximately to the first stage of the Goss procedure. In these prepared sheets, the grains are microscopic in size, and

Table 1 Weight percent chemical compositions of the silicon-iron sheet specimens used.

Si	C	Mn	P	S	Cu
3.50	0.005	0.16	0.013	0.023	0.30
1.43	0.010	0.11	0.016	0.032	0.16

we call them, for brevity, fine-grained sheet specimens in the following. Further, to obtain coarse-grained sheet specimens with the grain size of several mm to about 20 mm, the fine-grained 3.50 % silicon-iron sheets were cold-rolled 1 % of their original thickness and annealed at 850°C in pure dry hydrogen. Then, the fine grained as well as the coarse-grained sheet specimens were cold-rolled 0.5 to 10 % of their original thickness and annealed at 850°C in pure dry hydrogen. As mentioned in the introduction, this final procedure will be called, for brevity, the strain-anneal stage of our procedure hereafter. It is to be noted that, in the strain-anneal stage of our procedure, the cold rolling of fine grained 3.50 % silicon-iron sheet specimens were also made in 30, 60 and 90° angle directions to the initial rolling direction.

With these prepared sheet specimens, crystal grains were observed by the naked eye after being macro-etched with a 10 % aqueous solution of HNO₃ and the magnetic characteristics were measured by a magnetic permeameter. Also, the degree of the development of the Goss texture was determined by a torque magnetometer using discs 2 cm in diameter cut from the sheet specimens.

III. Experimental Results and Discussions

(I) Experiments on fine-grained sheet specimens

The macrostructures, after being subjected to the

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(1) N. P. Goss: T.A.S.M., 23 (1935), 511.

(2) J. M. Jackson: Japan Patent, No. 7504.

(3) Read at the General Meeting of the Japan Institute of Metals held at Tokyo, Japan, April 1948.

strain-annealing of fine-grained 3.50 and 1.43 % silicon-iron sheet specimens are shown in Photo. 1. Photo. 1 shows that the grain growth is most conspicuous at 2 % reduction for 3.50 % silicon and at 3 % reduction for 1.43 % silicon, respectively, and that the second growth maximum of the grain growth occurs at 6 to 7 % reduction for both silicon contents, the second maximum at 7 % reduction for 1.43 % Si being more significant.

$$L = -dE/d\theta = -K_1(2\sin 2\theta + 3\sin 4\theta)/8 + K_2(\sin 2\theta - 4\sin 4\theta - 3\sin 6\theta)/64, \quad (1)$$

where E is the ferromagnetic anisotropy energy density, θ is the angle between the direction of magnetization vector and the $[001]$ direction, and K_2 is the second cubic ferromagnetic anisotropy constant. For the actual comparison, of the measured and theoretical torque curves the first negative maximum, L_{m1} , and the ratio

Photo. 1 Effect of the reduction rate adopted in the strain-annealing stage of the author's procedure on the grain size of fine-grained 3.50 and 1.43 % Si-Fe sheet specimens. Numbers under photographs indicate the values of the reduction rate. Actual size.

The occurrence of two growth maxima seems to have been found by Stanley⁽⁴⁾ with some silicon-iron sheets as simply strain-annealed. It is to be noted that 3.50 % silicon-iron sheet specimens cold-rolled in directions different from the initial rolling direction in the strain-annealing stage of the author's procedure also showed quite similar macrostructures.

Next, the degree of development of the Goss texture in the sheet specimens may be determined from a comparison of the measured magnetic torque curves with the theoretical curves for the $(110)[001]$ single crystal with a positive cubic ferromagnetic anisotropy constant, K_1 , which is expressed as;

of the second positive maximum, L_{m2} , to L_{m1} have an important meaning. Because, for instance, when the texture of the sheet specimen has the (110) plane of grains coinciding with the rolling plane and the $[001]$ direction of grains centering at the two symmetrical directions nearby the rolling direction in the rolling plane, as is commonly seen in grain-oriented silicon-iron sheets, L_{m1} is lower than, but L_{m2}/L_{m1} is the same as, in the case of a single crystal, while the texture has the $[001]$ directions of grains coinciding with the rolling direction and the (110) planes of grains fluctuating about the rolling plane, L_{m1} is lower, but L_{m2}/L_{m1} is higher, than in a single crystal. Fig. 1 shows typical magnetic torque curves, measured after being subjected only to the first stage of the Goss procedure or of the author's procedure and after being subjected to the

(4) J. K. Stanley: Trans. AIME, 162 (1952), 116, cited by J. E. Burke et al in Progress in Metal Physics, 3 (1952), 242.

whole of our procedure, of fine-grained 3.50 % silicon-iron sheet specimens as well as the theoretical torque-curve of a (110) [001] single crystal of 3.0 % silicon iron for which L_{m1} and L_{m2}/L_{m1} are 16.5×10 dyne-cm/cm³ and 0.43, respectively⁽⁵⁾.

Fig. 1 Typical magnetic torque curves, measured after subsection only to the first stage of the Goss procedure (or of the author's procedure) (a) and after subsection to the author's entire procedure, (b) of fine-grained 3.50 % Si-Fe sheet specimens, and the theoretical curve of a (110) [001] single crystal of 3.0 % Si-Fe with positive K_1 (c).

In Fig. 2, the measured L_{m1} and L_{m2}/L_{m1} values are plotted against the reduction rate adopted in the strain-annealed stage of the author's procedure for fine-grained 3.50 and 1.43 % silicon-iron sheet specimens. It may be seen, from a comparison of Fig. 2 and Photo. 1, that

Fig. 2 Effect of the reduction rate used during the strain-anneal stage of the author's procedure on the magnetic torque maximum (negative), L_{m1} , and the ratio of the first and second magnetic torque maxima, L_{m2}/L_{m1} , for fine grained 3.50 and 1.43 % Si-Fe sheet specimens.

the L_{m1} value and the degree of grain growth as dependent on the reduction rate adopted in the strain-annealing stage of the author's procedure are alike, irrespective of the difference of silicon content. Thus, L_{m1} also shows two maxima at the reduction rate where the grain growth is maximum, whereas L_{m2}/L_{m1} shows a maximum value approximate to the theoretical value 0.43 for a (110) [001] single crystal of 3.0 % silicon iron at the reduction rate percentage where the first growth maximum locates, but it shows a minimum value at a reduction rate where the second growth maximum appears. Those results indicate that the Goss Texture is developed nearly perfectly at the reduction rate percentage where the first grain growth maximum appears, while, at the reduction rate corresponding to the second grain growth maximum, some other texture is also developed besides that of the Goss Texture.

Further, the torque measurements indicate that the initially fine-grained 3.50 % silicon-iron sheet specimens cold-rolled in directions different from the initial rolling direction in the strain-annealing stage of the author's procedure and showing macrostructures quite similar to those of the specimens cold-rolled in the initial rolling direction, as mentioned above, have a Goss texture which is already developed in the first stage of the Goss procedure, or of the author's procedure and has a distribution of grain orientation relative to the initial rolling direction, similar to that in the specimens made by cold rolling in the initial rolling direction.

Fig. 3 Effect of the reduction rate used during the strain-anneal procedure on maximum permeability, μ_{max} , and coercive force, H_c , for fine-grained 3.50 and 1.43 % Si-Fe sheet specimens.

(5) R. M. Bozorth: *Ferromagnetism*, (1951), p. 588.

It seems that Stanley⁽⁴⁾ attributes the two growth maxima observed with simply strain-annealed silicon iron sheets to the small nucleation activities of the primary recrystallization, but it may be concluded, from the above-mentioned macrostructural observations and magnetic torque measurements, that crystals developed at the first growth maximum in the initially fine-grained Goss-texture specimens have been grown from the Goss crystals with a small orientation fluctuation relative to the (110) [001] single crystal, possibly involving no primary recrystallization, and a crystal developed at the second growth maximum are a mixture of the Goss crystals and crystals belonging to certain textures recrystallized from the Goss texture.

Finally, the measured values of the maximum permeability, $\mu_{\max}$, and of the coercive force, H_c , are plotted against the reduction rates adopted in the strain-annealing stage of our procedure in Fig. 3. It may be seen from this figure that $\mu_{\max}$ shows two maxima, while H_c shows two minima, just corresponding to the two maxima observed with the grain growth (Photo. 1) and with Lm_1 (Fig. 2). Moreover, the magnetic hysteresis loops are highly rectangular compared with those of silicon-iron

0.35 mm thick, with various grain sizes below 3 mm. It has been found that the first grain growth maximum occurring at reduction rates between 2 and 3 % used for the strain-annealing stage of the author's procedure in fine-grained 3.50 and 1.43 % silicon-iron sheet specimens tends to disappear when the diameter of the grain is about 3 mm. This critical grain size is actually affected by the following conditions of the specimens before testing, thickness of specimen, amount of impurities, rolling conditions and texture of the initial specimen.

Then, detailed strain-annealing experiments were carried out using coarse-grained 3.50 % silicon-iron sheet specimens with a grain size much larger than 3 mm, which were prepared by 1 % cold reduction and subsequent annealing at 850°C in dry hydrogen. It was observed that, below 2 % cold reduction and at the annealing temperature of 850°C, the grain growth maximum did not appear and the texture was not changed very much, but, over 3 % reduction and at annealing temperatures higher than 850°C, new recrystallization occurred in the Goss crystals, as seen in Photo. 2, which show the grain structure before and after the strain

Fig. 4 Hysteresis loops of a 3.50 % Si-Fe sheet specimen prepared from a fine-grained sheet specimen by our procedure (the reduction rate adopted in the strain anneal stage of our procedure is 2 %) (solid line) as compared to the Goss procedure (broken line).

sheets with nearly the same degree of development of the Goss texture as prepared by the Goss procedure, as shown, for example, in Fig. 4.

Thus, based on the above-mentioned experimental results, we could obtain highly grain-oriented silicon-iron sheets with high permeability and low iron loss when using a low reduction rate of 2 % where the first grain growth maximum appeared.

(2) Experiments on coarse-grained sheet specimens

First the influence of the grain size on the basic materials which were subjected to the first stage of the Goss procedure (or of the author's procedure) on the grain growth obtained by the subsequent strain-annealing stage of the author's procedure was studied using cold-rolled and hot-rolled 1.2 to 3.5 % silicon-iron sheets,

Photo. 2 Effect of the strain anneal (3 % cold reduction and 850°C annealing) on coarse-grained 3.50 % Si-Fe sheet specimens. Photographs show the grain structures in the same region of the specimen before (a) and after (b) the strain anneal.

annealing of 3 % cold reduction and annealing at 850°C. These photographs were taken of a specimen etched slightly with a 10 % aqueous solution of HNO_3 using an optical apparatus similar to that described by Assmus et al.⁽⁶⁾ in which the directions of the incident light beam and of the observation direction were the same and

Fig. 5 Optical apparatus used for the observation of the textures of sheet specimens.

perpendicular to the specimen surface, as shown schematically in Fig. 5. We can see from these photographs that the Goss crystals showing a high-degree reflection

(6) F. Assmus, K. Detert and G. Ibe: *Z. Metallkde.*, 48 (1957), 345.

occupy a large portion of the specimen before the strain annealing, but, after this procedure, they are markedly decreased while the recrystallized dark regions increased.

The orientations of crystals in the recrystallized regions were found to be qualitatively similar to those of crystals found in 6 to 7 % strain-annealed fine-grained specimens. Thus, the measured Lm_1 and Lm_2/Lm_1 values before and after the strain-annealing procedure are shown in Table 2, which indicates that both Lm_1 , and Lm_2/Lm_1 , decrease and approach values corresponding to the second grain growth maximum observed with initially fine-grained sheet specimens (Fig. 2).

Table 2 A few examples of Lm_1 and Lm_2/Lm_1 values of initially coarse-grained 3.30 % Si-Fe sheet specimens before and after the strain-annealing procedure.

Specimen, No.	Lm_1 (10 dyne-cm/cm)		Lm_2/Lm_1	
	Before	After	Before	After
1	10.3	7.6	0.38	0.12
2	14.4	9.3	0.41	0.11
3	9.1	7.1	0.34	0.16

These facts may be summarized as follows:

(i) The recrystallization takes place in Goss crystals in the second grain growth maximum of 6 to 7 % cold-rolled fine-grained sheet specimens as well as in more

than 3 % cold-rolled coarse-grained sheet specimens.

(ii) The difference in the above reduction rate may be attributed to the fact that in fine grained sheet specimens, the first grain growth tendency shifts the primary recrystallization to a 6 to 7 % reduction rate, revealing the second grain growth maximum.

It was noticed in (1) that the hysteresis loops of the specimens produced by our strain-annealing procedure were highly rectangular. In Figs. 6 and 7, the values of the residual induction Br , and coercive force H_c , measured on coarse-grained sheet specimens are functions of the reduction rates and of the annealing temperature adopted in the strain-annealing stage of our procedure.

Fig. 7 Effect of the reduction rate on the residual induction of the strain-annealed coarse-grained 3.50 % Si-Fe sheet specimens.

Fig. 6 (a) shows the residual induction Br for the maximum induction of 15 kilo-Gauss as dependent on the annealing temperature as compared with that of the Goss processed specimens. In coarse grained specimens, Br increases rapidly with increasing annealing temperature, passes a maximum value and then decreases to some extent whereas that of the Goss processed specimen decreases uniformly with rising annealing temperature. On the other hand, as seen from Fig. 6 (b), H_c decreases with increasing annealing temperature for both specimens. The decrease of Br from a maximum value in coarse-grained specimens (Fig. 7) corresponds to the appearance of recrystallization.

IV. Summary and Conclusions

The grain growth after being light cold rolled at rates of 3.50 and 1.43 % of silicon-iron sheets with Goss texture was investigated, using both fine-grained and coarse-grained sheet specimens.

Sheet specimens prepared as above, were macro-etched with a 10 % aqueous solution of HNO_3 and visually inspected for crystal grains and the magnetic characteristics were measured by a magnetic permeameter. Also the degree of the development of the Goss texture was determined by a torque magnetometer.

In originally fine-grained sheet specimens, two maxima of the grain growth were found in rolling reduction ranges between 0.5 to 2 % and from 6 to 7 %, respectively.

It may be concluded, from the above facts, that at the first grain growth maximum, the resultant crystals have been grown from the Goss crystals with small orientation fluctuations in the specimens, and possibly the primary recrystallization has not occurred and that, at the second growth maximum, the resultant crystals have been developed by primary recrystallization from the Goss crystals.

Fig. 6 Effect of the annealing temperature on the residual induction, Br , and coercive force, H_c , of strain-annealed coarse-grained 3.50 % Si-Fe sheet specimens as compared with 3.50 % Si-Fe sheet specimens prepared by the Goss procedure (60 % cold rolled).

In originally coarse-grained specimens, the first grain growth maximum was not found and it may be considered that, at about 3 % reduction, the primary recrystallization occurs in the Goss crystals and the resulting textures corresponds to those found in the second grain growth maximum in initially fine grained specimens.

It may be concluded from the observed magnetic characteristics, that the optimum point to obtain highly oriented silicon-iron sheets with high permeability and low hysteresis loss is at the first grain growth maximum found with initially fine-grained sheet specimens.

Rectangular shaped hysteresis loops were found with

initially fine-grained as well as with initially coarse-grained sheet specimens subjected to the strain-annealing procedure.

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