June 9, 1964 SATORU TAGUCHI ETAL 3,136,666
METHOD FOR PRODUCING SECONDARY RECRYSTALLIZATION
GRAIN OF CUBE TEXTURE
Filed Jan. 23, 1961 2 Sheets-Sheet 1

FIG. I

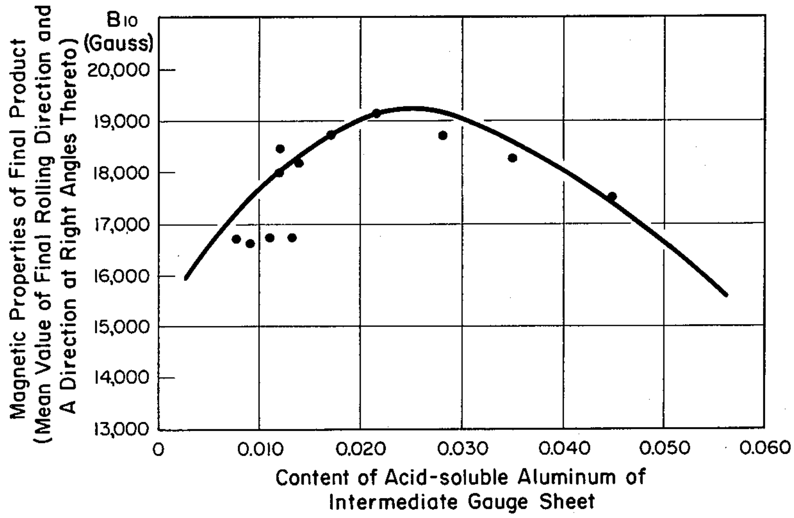

FIG. 2-H SC Texture

(IOO) Pole Figure

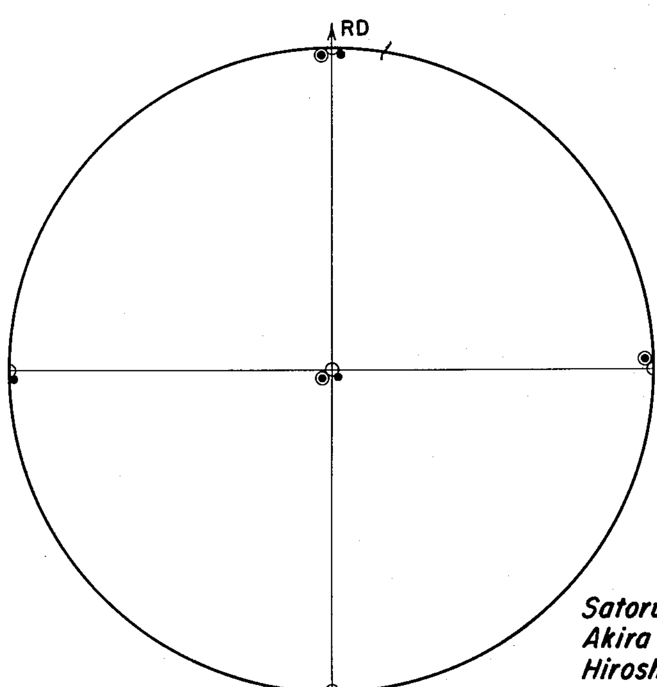

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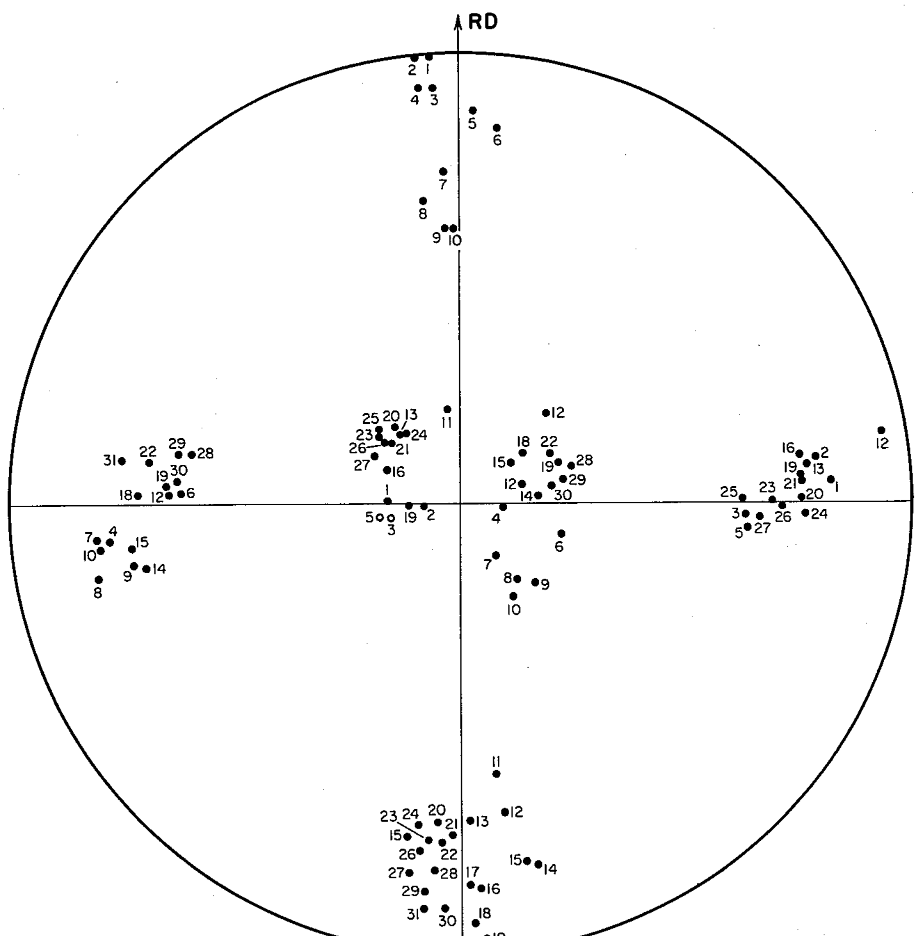
FIG. 2-G PC Texture

(100) Pole Figure



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United States Patent Office

3,136,666
Patented June 9, 1964

1

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METHOD FOR PRODUCING SECONDARY RECRYSTALLIZATION GRAIN OF CUBE TEXTURE

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Filed Jan. 23, 1961, Ser. No. 84,421
Claims priority, application Japan Jan. 27, 1960
2 Claims. (Cl. 148—111)

The present invention relates to a method for producing a cube-oriented silicon steel sheet having a coarse large grain in the so-called (100) [001] orientation, that is, having an easy direction or axis of magnetization [001] in two directions mutually perpendicular on the rolling plane and also having (100) plane in the rolling plane.

Coarse large grains are obtained from the so-called secondary recrystallization resulting by the selective growth of primary recrystallization grains produced by an anneal, which is fully described hereinafter. In the production of a grain-oriented silicon steel sheet in which the crystal lattice consisting of each grain is required to be aligned almost in the same orientation, the formation of secondary recrystallization grains has been one of the necessary steps. This phenomenon is called "secondary recrystallization" or "grain coarsening" in metallurgy, and characterized by the fact that the selective growth of a grain with a specified orientation only takes place.

Where there is lack of a factor for developing secondary recrystallization in the silicon steel sheet, it is usual that the growth of primary recrystallization grain resulting from the anneal occurs, which is called "normal grain growth."

However, if the gauge of a silicon steel sheet is thick in spite of its preferred orientation, when it is applied for an iron core for use in a transformer or other electric appliances, eddy current losses become high, which, therefore, results in a high core loss. In order to eliminate this disadvantage, we contemplate the provision of a magnetic sheet reduced further in thickness. As a method for obtaining a thin sheet, there are two methods of making it, either chemical or mechanical, such as, grinding or cold rolling (and anneal). The latter is adopted becouse it is economical as well as commercial. In general, however, the complete reproduction of the preferred orientation of grains which constitutes an intermediate gauge sheet by the cold rolling and annealing procedure is rather difficult.

A principal object of the invention is to provide a method for producing a thin gauge silicon steel sheet which comprises reducing the intermediate gauge sheet having coarse large grains of the (100) [001] orientation using as a starting material by cold-rolling said intermediate gauge silicon steel sheet in the direction of substantially [001] orientation and reproducing the secondary recrystallization grains of the (100) [001] orientation in the cold-rolled steel sheet by subsequently annealing the latter. To this end, the present invention has for its important feature to have the above-mentioned intermediate gauge steel sheet containing 0.010 to 0.045% acid-soluble aluminum, a part of which consists of aluminum nitride, AlN.

2

Referring to the accompanying drawings:

FIG. 1 is a diagram showing the relation between the content of an acid-soluble aluminum of the intermediate gauge sample and the magnetic properties of the sample after the final anneal.

FIG. 2 is the (100) pole figure showing the grain orientation of the sample after the final anneal.

The invention will be fully described hereinbelow.

An intermediate gauge silicon steel sheet having a coarse large grain in the (100) [001] orientation, a starting material for carrying out the invention, can be produced by various methods. As a preferred example, however, a method disclosed in our copending application Serial No. 844,450, filed October 5, 1959, now abandoned, will be explained. According to the method disclosed in the above copending application Serial No. 844,450, a hot rolled silicon steel stock containing 2.0–4.0% Si and 0.010–0.050% Al is cold rolled in one direction with a reduction of 30% to 60% in thickness, again cold rolled in the other crossing direction thereto within a deviation of $\pm 20°$ in angle with a reduction of 20% to 50% in thickness, and thereafter subjected to an intermediate anneal at the maximum holding temperature within the range of 850° to 1200° C. By this method, an intermediate gauge silicon steel sheet, a starting material, can be obtained. It can be seen that the above intermediate gauge silicon steel sheet consists of the secondary recrystallization grain in the (100) [001] orientation.

Now, the effect of the acid-soluble aluminum, the most significant feature of the invention, will be described. Referring to Table 1, A, B and C are hot rolled silicon steel stock of 3.0 mm. gauge containing three ingredients, respectively, and each of these starting materials is cold rolled in the same rolling direction as that of hot rolling with a reduction of 40% in thickness in accordance with the teachings disclosed in the copending application Serial No. 844,450, again cold rolled in a direction at right angles thereto with a reduction of 40% to produce an intermediate gauge 1.08 mm. sheet, then subjected to a decarburizing anneal at the temperature of 800° C. for a period of 10 minutes, and thereafter to an intermediate anneal in an atmosphere of H_2 containing 50% N_2 at the maximum holding temperature of 1150° C. in order to obtain an intermediate gauge silicon steel sheet consisting of secondary recrystallization grains in the complete (100) [001] orientation. The thus produced intermediate gauge silicon steel sheet is subjected to an additional anneal to effect formation of various contents of acid-soluble aluminum contained in the metal, then again cold rolled in a direction corresponding substantially to the [001] grain direction with a reduction of 72% in thickness to the final gauge, and finally subjected to the final box anneal at the highest holding temperature of 1200° C. for a period of 20 hours. In addition, Table 1 clearly indicates the relation between the content of acid-soluble aluminum contained in the above intermediate gauge sheet after the above additional anneal, the state of recrystallization after the final anneal and magnetic properties of final products.

TABLE 1

Sample No.	Analysis, by weight percent			Condition of additional anneal after intermediate anneal	Analysis after additional anneal, by weight percent				Magnetic properties of final product		State of recrystallization in final anneal
	Si	Total Al	Acid soluble Al		Acid soluble Al	N as AlN	Al as AlN	Al as AlN/sol. Al (percent)	B_{10}	W 15/50	
A 1	2.98	0.019	0.014	Intermediate anneal only	0.012	0.0050	0.0096	80	18,050 18,000	1.18 1.20	SC
A 2				1200° C., 1 hr. in H_2	0.010	0.0040	0.0077	77	17,550 17,800	1.40 1.25	SC
A 3				1200° C., 20 hr. in H_2	0.008	0.0010	0.0019	24	16,700 16,650	1.56 1.52	PC
B 1	3.05	0.034	0.030	Intermediate anneal only	0.022	0.0095	0.0183	60	19,200 19,100	1.01 1.02	SC
B 2				1200° C., 1 hr. in H_2	0.014	0.0060	0.0116	83	18,340 18,150	0.98 1.05	SC
B 3				1200° C., 20 hr. in H_2	0.009	0.0021	0.0041	46	16,800 16,300	1.50 1.58	PC
B 4				1200° C., 20 hr. in vacuum 2×10^{-5} mm. Hg.	0.011	0.0000	0.0000	0	16,750 16,700	1.49 1.51	PC
C 1	3.04	0.050	0.041	Intermediate anneal only	0.035	0.0139	0.0268	77	18,200 18,250	1.16 1,15	SC
C 2				1200° C., 1 hr. in H_2	0.028	0.0065	0.0125	45	18,700 18,700	1.10 1.05	SC
C 3				1200° C., 20 hr. in H_2	0.017	0.0032	0.0062	37	18,800 18,750	1.01 1.08	SC
C 4				1200° C., 60 hr. in H_2	0.012	0.0008	0.0015	13	18,450 18,400	1,15 1.15	SC
C 5				1200° C., 20 hr. in vacuum 2×10^{-5} mm. Hg.	0.013	0.0000	0.0000	0	16,700 16,700	1.52 1.51	PC

Remarks:

1. B_{10} is magnetic induction at 10 oersteds and its unit is gauss.
2. W 15/50 is iron core loss at 15,000 gauss measured at 50 cycles and its unit is watt/kg.
3. In magnetic properties of sample, the upper value is that of final rolling direction and the lower that of a direction at right angles thereto.
4. PC is primary recrystallization texture and SC is secondary recrystallization texture.
5. Sol. Al is acid-soluble aluminum.
6. Al as AlN of analysis is calculated based on N as AlN.

The following will become clear from Table 1. That is to say, in case intermediate gauge silicon steel sheets A_3, B_3 and B_4 containing acid-soluble Al less than 0.010% were used as starting materials, a primary recrystallization structure was produced after the final annealing. It had orientations somewhat deviating from (100)[001] orientations and found by calculation to be quadruplet {113}<301> orientations (a typical example of which is shown in FIGURE 2G). Its magnetic characteristic (magnetic induction B_{10}) was of a low value of about 16,600 gausses in each of the rolling direction and the direction at right angles thereto. On the other hand, in case intermediate gauge silicon steel sheets A_1, A_2, B_1, B_2, C_1, C_2, C_3 and C_4 containing Al more than 0.010% were used as starting materials, after the final annealing, a secondary recrystallization structure was produced. Its crystal orientations were substantially perfect (110) [001] orientations (a typical example of which is shown in FIGURE 2H). Its magnetic induction B_{10} was more than at least 17,000 gausses in each of the rolling direction and the direction at right angles thereto and the highest value was shown to be more than 19,000 gausses.

FIGURE 1 shows the relation between the content of acid-soluble Al in the intermediate gauge silicon steel sheet and the magnetic property after the final annealing. It is found from FIGURE 1 that the content of acid-soluble Al in an intermediate gauge silicon steel sheet required in order that the magnetic induction B_{10} of the steel sheet after the final annealing may show a value above 17,000 gausses in each of the cold-rolling direction and the direction at right angles thereto must be within a range of 0.010 to 0.045%. This is the first requirement that the intermediate gauge steel of the present invention should have.

Now, the content of AlN shown in Table 1 will be described. It is known that the analysis of acid-soluble aluminum determined by the ordinary chemical analysis contains AlN. Where the intermediate gauge silicon steel sheet of samples B and C shown in Table 1 is subjected to the additional anneal in the high vacuum of 2×10^{-5} mm. Hg, AlN is not present in spite of the fact that the acid-soluble aluminum is contained above 0.010%. In this case, the grain growth texture of the primary recrystallization grain (PC texture) is formed by the final anneal, and the secondary recrystallization texture (SC texture) in the (100)[001] orientation, the object of the invention, is not achieved. After an extensive study of experimental results, we have discovered that the presence of AlN required for the development of the SC texture does not depend upon its absolute amount, but upon the relative amount in connection with the acid-soluble aluminum. That is to say, at least more than 5% of acid-soluble Al must be present in the form of AlN. This is the second requirement of the intermediate gauge silicon steel sheet to be used as the starting material in the present invention. It is to be understood however, that the above finding holds true as long as the content of the acid-soluble aluminum remains in the range of 0.010 to 0.045% which meets the first essential requirement of the invention. The silicon steel sheet of the present invention must meet both of the above-mentioned first and second requirements. That is to say, only when these two requirements are fulfilled, the silicon steel sheet of the intermediate gauge having crystal grains of the (100)[001] orientation may completely reproduce the secondary recrystallization grains of (100) [001] orientation after cold-rolled and subsequently annealed.

The above finding applies to any intermediate gauge silicon steel sheet produced by any other method than disclosed in our co-pending application Ser. No. 854,450, now abandoned. But, also in such an intermediate gauge silicon steel sheet, it is requisite that the acid-soluble Al is contained in the range of 0.010 to 0.045% and at least more than 5% of said Al is present in the form of AlN to attain the object of the present invention.

Further, the Si content in the silicon steel sheet of the intermediate gauge of the present invention shall be 2 to 4%.

When Si exceeds 4%, the steel will become brittle and difficult to roll while cold. When Si is less than 2%, an α-γ transformation will occur when annealing after cold-rolled and the intended secondary recrystal grains of (100)[001] orientations will not be produced. Further, if Si is low, the electric resistance will be low and there will be a disadvantage that the core loss value will increase. Therefore, Si is defined to be 2 to 4%.

Next, the intermediate gauge sheet is finally cold rolled in the substantially [001] direction of the grain with a preferred reduction in thickness in the range of 50 to 84%. If the reduction in thickness is either below 50% or above 84%, the secondary recrystallization grain in the (100)[001] orientation, the object in view, is not fully developed. Thus, the magnetic induction of the final product is considerably lowered because of many recrystallization grains in other orientations. We recommend a reduction in thickness of 70% or thereabouts as the most preferred reduction of the final cold rolling step.

The thus produced final gauge steel sheet is subjected to the final anneal. To obtain the desired secondary recrystallization grain in the (100)[001] orientation, the final anneal should be carried out in a neutral or reducing gas atmosphere at a temperature of above 1000° C. A complete grain growth is not obtained at a temperature of below 1000° C., nor effective at a temperature of above 1300° C. We prefer the temperature range of 1000° to 1300° C. as the most suitable one for the final anneal.

As clearly described in the foregoing, the feature of the present invention comprises subjecting the intermediate gauge steel sheet of the (100)[001] grain orientation containing 0.010 to 0.045% acid-soluble aluminum, at least more than 5% of which acid-soluble Al is present in the form of AlN, to the cold rolling step in the direction corresponding almost to the [001] direction of the grain to the final gauge, and then to the final anneal to produce a cube-oriented silicon steel sheet consisting of the secondary recrystallization grain (SC texture) in the (100)[001] orientation.

If the intermediate gauge sheet consisting of the (100)[001] grain orientation does not contain acid-soluble aluminum and AlN in the above specified range, the PC texture produced by the final anneal deviates considerably from the (100)[001] orientation as shown at G in FIG. 2. In addition, the magnetic properties including magnetic induction and iron core loss are inferior as illustrated in Table 1.

Different magnetic properties of final products resulting from different contents of acid-soluble aluminum and AlN will develop a significant difference in the magnetic properties when they are applied for iron core for electric equipment.

Example 1

Two kinds of single crystals G and H are prepared, each of which contains acid-soluble aluminum, nitrogen as AlN (as obtained by the calculation

$$\text{Al as AlN(percent)} = \text{N as AlN(percent)} \times \frac{\text{atomic weight of Al (27)}}{\text{atomic weight of N (14)}}\Big)$$

0.007%, 0.0009% and (0.0017%) and 0.025%, 0.0100% and (0.0193%) respectively, with the substantially complete (100)[001] orientation, about 30 mm. diameter and 1.08 mm. thick. Each of these samples is cold rolled with a reduction in thickness of 70% in the [001] direction and finally annealed at the maximum holding temperature of 1200° C. for a period of twenty hours. As a result, sample G is an aggregate of fine crystals of about 1 to 2 mm. diameter resulting from the grain growth of primary recrystallization grain and its orientation deviates fairly from (100)[001] as at G shown in FIG. 2 while, on the other hand, sample H produces again the secondary recrystallization grain about 20–30 mm. diameter with the orientation of a complete (100)[001] type as at H of FIG. 2. Further, the figures 1, 2, 3 . . . of FIG. 2 are the numerals of representative grains of sample G.

Example 2

A hot rolled silicon steel stock containing 3.02% Si, 0.033% Al, and 0.029% acid-soluble aluminum at gauge 3.00 mm. is cold rolled with a reduction in thickness of 40% in the same direction as that of hot rolling to obtain the sheet 1.8 mm. thick, and then cold rolled with a reduction in thickness of 40% in a direction substantially at right angles to that of the primary cold rolling with the result that a cold rolled silicon steel sheet of an intermediate gauge 1.08 mm. is obtained. Subsequently, the intermediate gauge sheet is annealed in the atmosphere of a mixture of H_2 and N_2 in the proportion of 50 to 50 by volume at the maximum holding temperature of 1150° C. for a period of 15 hours in order to develop an easy axis of magnetization [001] in the above two directions of cold rolling, that is, the secondary recrystallization grain in the so-called (100)[001] orientation, in which the content of the acid-soluble aluminum is maintained to 0.021% including 0.0094% N as AlN and 0.0182% Al as AlN (calcu.).

Subsequently, the intermediate gauge sheet is again cold rolled with a reduction in thickness of 72% in a direction almost corresponding to one of the above two cold rolling directions to the final gauge, then subjected to decarburization by annealing it in a wet hydrogen gas at the temperature of 800° C. for a period of three minutes, and to the final anneal at the maximum holding temperature of 1200° C. for a period of 20 hours to produce again the secondary recrystallization grain in the (100) [001] orientation, which results in an excellent magnetic property.

The results of magnetic test conducted on Epstein sample taken from the steel sheet in the final rolling direction and also in a direction at right angles thereto and then subjected to a strain relief anneal are shown in Table 2, showing also an excellent cube-oriented silicon steel sheet.

Table 2

	B_{10}, gauss	W 15/50 (w./kg.)
Final rolling direction	19,150	0.99
Direction at right angle to final rolling direction	19,100	1.06

We claim:

1. Process for producing thin gauge sheets of double-oriented silicon steel of crystal grain orientation (100) [001], comprising subjecting an intermedite gauge silicon steel sheet consisting of crystal grains of predominantly (100)[001] orientation at least once to
   (1) cold rolling in a direction substantially coinciding with the [001] direction with a reduction rate of about 50 to 84%, and
   (2) annealing at a temperature of about 1000 to 1300° C., wherein said intermediate gauge starting material contains about 2.0 to 4.0% by weight of Si, and the steel sheet after each non-final annealing step contains about 0.010 to 0.045% by weight of acid-soluble Al, said acid-soluble Al consisting by more than 5% of AlN calculated according to the formula

$$100 \times \frac{\text{Al as AlN (based by percent on weight of steel)}}{\text{acid-soluble Al percent based on weight of steel)}} \geq 5 \text{ (percent)}$$

2. Process for producing thin gauge sheets of double-oriented silicon steel of crystal grain orientation (100)[001] comprising
   (1) cold rolling intermediate gauge silicon steel sheet consisting of crystal grains of predominantly (100) [001] orientation in a direction substantially coinciding with the [001] direction with a reduction rate of 50 to 84%, said steel sheet containing about 2.0 to 4.0% by weight Si and about 0.010 to 0.045% by weight of acid-soluble Al, said acid-soluble Al

consisting by more than 5% by weight of AlN calculated according to the formula

$$100 \times \frac{\text{Al as AlN (percent based on weight of steel)}}{\text{acid-soluble Al (percent based on weight of steel)}} \geqq 5 \text{ (percent)}$$

and

(2) annealing said rolled sheet at a temperature of 1000° to 1300° C., thereby reproducing crystal grains of (100) [001] orientation.

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