

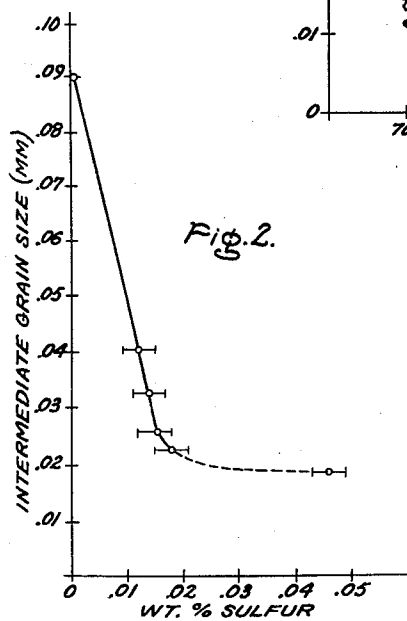
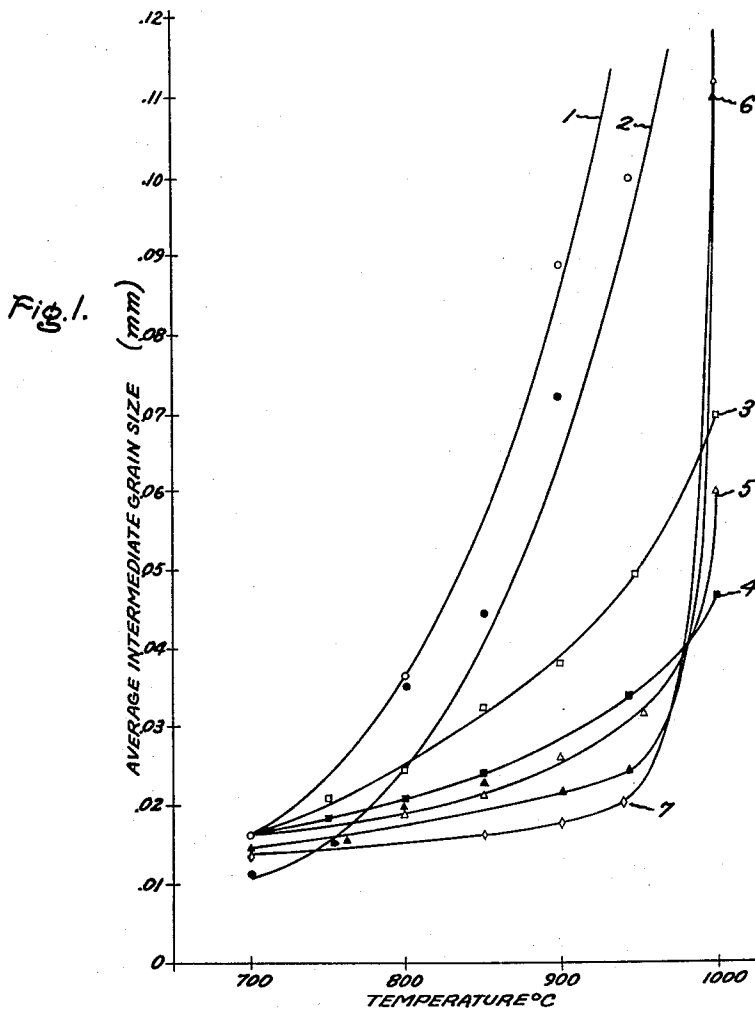
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METHOD FOR PRODUCING GRAIN-ORIENTED SILICON STEEL

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METHOD FOR PRODUCING GRAIN-ORIENTED SILICON STEEL

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5 Claims. (Cl. 148—111)

This invention relates to polycrystalline magnetically "soft," rolled sheet metal composed principally of an alloy of iron and silicon and more particularly to a process for manufacturing such materials wherein a high percentage of the grains comprising the material are each caused to have their crystal space lattices arranged in a substantially identical relationship to the plane of the sheet and to a single direction in the plane of the sheet.

The sheet materials to which my invention is related are usually referred to in the art as "electrical" silicon steel or, more properly, silicon-iron, composed primarily of iron alloyed with about 2.5 to 4.0 percent silicon and containing relatively minor amounts of impurities, such as sulfur, manganese, phosphorus, and preferably, a very low carbon content. Such alloys crystallize in the body-centered cubic crystallographic system at room temperature. As is well known, this refers to the symmetrical distribution or arrangement which the atoms forming the individual crystals or grains assume in such materials. In these materials, the smallest prism possessing the full symmetry of the crystal is termed the unit cell and is cubic in form. This unit cube is composed of nine atoms, eight arranged at the corners of the unit cube with the remaining atom positioned at the geometric center of the cube. Each unit cell in a given grain or crystal in these materials is substantially identical in shape and orientation with every other unit cell comprising the grain.

The unit cells or body-centered unit cubes comprising these materials each have a high degree of magnetic anisotropy with respect to the crystallographic planes and directions of the unit cube and hence, each grain or crystal comprising a plurality of such unit cells exhibits a similar anisotropy. More particularly, crystals of the silicon-iron alloys to which this invention is directed are known to have their direction of easiest magnetization parallel to the unit cube edges, their next easiest direction of magnetization perpendicular to a plane passed through diagonally opposite parallel unit cube edges and their least easiest direction of magnetization perpendicular to a plane passed through a pair of diagonally opposite atoms in a first unit cube face and a single atom in the unit cube face which is parallel to the first face. As is well known, these crystallographic planes and directions are conventionally identified in terms of "Miller Indices," a more complete discussion of which may be found in "Structure of Metals," C. S. Barrett, McGraw-Hill Book Company, New York, New York, 2nd edition, 1952, pp. 1-25 and will be referred to as, respectively, the (100) plane and the [100] direction, the (110) plane and the [110] direction, and the (111) plane and the [111] direction.

It has been found that the silicon-iron alloys may be fabricated by unidirectional rolling and heat treatment to form sheet or strip material composed of a plurality of crystals or grains, a majority of which have

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their atoms arranged so that their crystallographic planes have a similar or substantially identical orientation to the plane of the sheet or strip and to a single direction in said plane. This material is usually referred to as "oriented" or "grain-oriented" silicon-iron sheet or strip and is characterized by having 50 percent or more of its component grains oriented so that four of the cube edges of the unit cells of such grains are substantially parallel to the plane of the sheet or strip and to the direction in which it was rolled and a (110) crystallographic plane substantially to the plane of the sheet. It will thus be seen that the so-oriented grains have a direction of easiest magnetization in the plane of the sheet in the rolling direction and the next easiest direction of magnetization in the plane of the sheet in a transverse to rolling direction. This is conventionally referred to as a "cube on edge" orientation or the "(110) [001] texture." In these polycrystalline sheet or strip materials, it is desirable to have as high a degree of grain orientation as attainable, preferably more than 70 percent, in order that the magnetic properties in the plane of the sheet and in the rolling direction may approach the maximum attained in single crystals in the [100] direction.

In actual steel mill practice, these alloys are cast into ingots hot worked, usually by rolling, to less than 0.15" thick sheets or strips called "band" and then subjected to varying schedules of usually unidirectional cold rolling and heat treatment to produce the final, oriented sheet or strip, usually about 0.010 to 0.015" in thickness. Unfortunately, the degree of final orientation produced by prior known practices has been quite variable and has necessitated magnetic testing of samples cut from each such finished strip to determine its degree of orientation. Since such materials having relatively low degrees of orientation are undesirable for many electrical applications and hence are not as useful as materials having higher degrees of orientation. In previous practices, it is not uncommon that oriented sheet or strip prepared from batches or "heats" having substantially identical compositions and processed in substantially identical fashion will yield finished strips or sheets only about 60 percent or less of which have 70 percent or more of their grains oriented as desired.

As pointed out in my copending application, Serial No. 631,889, filed concurrently herewith and assigned to the same assignee, it is important that the grain size of these materials be controlled at an intermediate stage of the fabrication in order to assure the consistent production of highly oriented final sheet or strip material. Furthermore, it has been found that during the final heat treatment of these materials, a phenomenon referred to as "secondary recrystallization" must take place in order for well developed (110) [001] textures to develop. I have discovered that there is a critical relationship between the amount, kind and disposition of impurities present and the ability of these materials to respond to heat treatment to cause secondary recrystallization.

A principal object of my invention is the provision of a method of fabrication of such silicon-iron alloys to insure that the highest attainable degree of grain orientation may be consistently produced in the final sheet or strip material. Other and specifically different objects of my invention will become apparent to those skilled in the art from the following detailed disclosure.

Briefly stated, in accordance with one aspect of my invention, I have discovered that critical amounts of finely dispersed, relatively stable inclusions or impurities are essential to the degree of final texture which may be attainable in these materials and that by controlling these impurities and the grain size at intermediate stages

of fabrication that higher degrees of texture and higher final annealing temperatures and therefore shorter annealing times may be successfully employed resulting in greater economy in manufacture.

My invention will be better understood from the following detailed description taken in connection with the accompanying drawing and its scope will be pointed out in the appended claims.

In the drawings, Fig. 1 is a graphical representation of the effect of temperature and impurity composition on intermediate grain size and Fig. 2 is a graphical representation of the relationship between impurity and intermediate grain size under an exemplary given annealing treatment.

As pointed out in greater detail in my copending application previously referred to, the grain size of these materials at an intermediate stage of fabrication has an important and controlling relationship upon the final degree of texture attainable. More particularly, these materials are conventionally produced by hot reducing ingots to sheet or strip-like bodies, usually less than 0.150" in thickness, called "band," usually by rolling operations. These materials are then subjected to cold reduction by rolling with appropriate intermediate heat treatment to final thickness, for example, about 0.01 to 0.015" thickness. This cold rolled material is then conventionally given a decarburizing heat treatment and a texture developing anneal. The cold rolling schedule employed is usually one in which the cold reduction is accomplished in at least two steps, separated by an annealing treatment. It is essential, as pointed out in my copending application, to control the grain size of the annealed strip before cold reducing the strip to final thickness. This grain size is referred to as the "intermediate" grain size, and should be maintained between an average measured grain size of about 0.010 and 0.030 mm.

It has been found that the amount of cold reduction necessary to produce this critical intermediate grain size is at least 40 percent in thickness during the penultimate cold rolling sequence and that a cold reduction of at least 40 percent is necessary in the ultimate or final cold rolling sequence in order to develop the desired degree of orientation.

During the texture developing heat treatment of these highly oriented materials, it has been observed that the recrystallization of the worked material occurs in two general ways. First, the material develops an equiaxed relatively uniform fine grain structure, usually referred to as the "primary" recrystallization structure. These grains have essentially a random orientation, in other words, a relatively few have the desired texture while the majority have other, specifically different orientations. As the heat treatment continues, these primary grains tend to grow somewhat, however, grains having the (110) [001] orientation grow at a much greater rate than the primary matrix grains and consume their less favorably oriented neighboring grains. These secondary grains are easily identifiable since they become during their development, up to millions of times as large as the primary grains. It has been found that unless the secondary recrystallization phenomenon occurs during this heat treatment, well developed or "strong" (110) [001] textures are not developed.

As will become apparent from the following disclosure, I have discovered that certain critical amounts of impurities are necessary in these materials in order to produce and control the desired intermediate grain size and the final degree of texture finally developed.

In order to particularly point out my invention, the following specific examples are set forth. A number of alloys were prepared by vacuum melting the compositions reproduced in Table I. Commercial electrolytic iron and 99.95 percent pure silicon were used as the basic materials. Certain selected impurities were added as electrolytic manganese and iron sulfide.

Table I

Alloy	Composition, weight percent ¹				
	Si	Mn	S	P	C
1	3.34	0.000	0.005	-----	0.003
2	3.34	.000	.000	-----	-----
3	2.70	.098	.012	0.003	.003
4	2.34	.040	.014	.003	.003
5	2.53	.038	.015	-----	.003
6	2.98	.046	.018	.002	.003
7	2.84	.110	.046	-----	.006

¹ Balance substantially all iron.

It will be noted that alloys listed in the foregoing table were melted and cast as 6 pound ingots by melting the iron under vacuum, deoxidizing the melt with hydrogen for 30 minutes, adding the silicon and selected impurities, permitting the melt to solidify, pumping the hydrogen out, recreating a vacuum, and remelting and casting the alloys under the vacuum. All the compositions listed in Table I are the result of chemical analyses performed upon the ingots.

The ingots resulting from casting these alloys were forged and drawn at temperatures between 800° C. and 1000° C. to 2" x 1" cross-section bars. These bars were then hot-rolled to from 0.2" to 0.4" in thickness and then cold-rolled to about 0.100" thick and annealed to obtain a characteristic equi-axed grain structure. These annealed bands were cold reduced from 0.100 to 0.028" intermediate thickness and separate portions were annealed at different temperatures for 15 minutes each in a protective atmosphere. The grain size of each of the portions was determined, the grain size versus temperature plotted in Fig. 1, and the portions were cold rolled to 0.014" thickness strip. Portions of each so-reduced strip were annealed in a protective atmosphere at different temperatures for 1 hour each and the primary grain size of each was measured and is reproduced in Table II.

Table II

GROUP I.—INTERMEDIATE ANNEALING TREATMENTS FROM 700-1,000° C.

Alloy	Primary matrix grain size after final anneal, mm.		
	700° C.	945° C.	1,000° C.
1	0.0142	0.05	0.15
2	.0133	.06	-----
3	.0156	.086	.06

GROUP II.—INTERMEDIATE ANNEALING TREATMENTS FROM 700-930° C.

4	0.018	0.030	0.032
5	.0148	.026	.030
6	.0135	.024	.032
7	.0125	.020	.025

GROUP III.—INTERMEDIATE ANNEAL AT 1,000° C.

4	0.0188	0.0325	0.040
5	.0198	.032	.036
6	.0255	.030	.038
7	.0143	.036	.032

The remaining portions of each cold reduced strip were annealed in a temperature gradient furnace. In this anneal these strips were arranged in an annealing furnace so that the temperature at one end of each strip was maintained at 850° C. and the temperature gradually increased along the length of each strip to 1145° C. at the other end of each strip. After these strips have been annealed for a sufficient time to insure complete recrystallization, they were removed, etched, and subjected to macroscopic grain structure examination.

As will be observed, with particular reference to Fig. 1 and Table I, a temperature range for annealing alloys

1 and 2 which contain 0.005 percent and no sulfur respectively, in order to produce intermediate grain sizes between 0.01 and 0.030 mm. is very narrow. As the amount of sulfur and manganese are increased, this critical range of grain size is obtainable over much wider temperature ranges. For example, alloy 7 with a sulfur content of 0.046 percent and a manganese content of 0.110 percent may be annealed under these conditions at temperatures as high as about 975° C. without causing excessive grain growth. Furthermore, as will be later seen, alloys 1 and 2 are incapable of developing any appreciable degree of the desired structure.

During the final annealing treatment, two stages of recrystallization occur during which time in the high orders of textures are developed. The cold-worked material undergoes an initial or primary recrystallization in which a great number of relatively small grains of varying orientations develop, then if the material contains a sufficient amount of sulfur, probably present as a manganese sulfide inclusion, certain of these grains grow at the expense of others, forming very large "secondary" grains. These secondary grains may be seen by a macroscopic examination.

An examination of the strips annealed at the various intermediate temperatures as grouped in Table II and given the indicated final annealing treatment, revealed the secondary recrystallization characteristics of each specimen. It was found that one group, namely strips from alloys 1, 2 and 3 showed no evidence of a secondary recrystallization phenomenon regardless of the intermediate annealing temperature employed. Strips from alloys 4, 5, 6, and 7 revealed secondary recrystallization in samples annealed at temperatures between 945° and 1000° C. Strips from alloys 4, 5, 6 and 7 which had been given an intermediate anneal at 1000° C. did not exhibit any secondary recrystallization. It is noted, as shown in Table II that there is a relationship between the primary grain size and the final annealing temperature with the tendency to exhibit the secondary recrystallization phenomenon in that the primary matrix grain size for alloys 1, 2 and 3 increases more rapidly with respect to temperature than do alloys 4, 5, 6 and 7.

The strips which have been annealed in the temperature gradient furnace were inspected and it was found that secondary recrystallization was exhibited in alloys 4, 5 and 6 in the temperature range between about 875° C. and 975° C. whereas in alloy 7 this phenomenon was found to occur between about 875° C. and 1100° C.

It is apparent that a minimum amount of relatively stable inclusions present as a second phase dispersion are necessary to inhibit the growth of primary matrix grains having orientations other than the (110) [001] during the final annealing treatment whereby the (110) [001] texture grains may grow at the expense of others. In addition, this minimum amount of impurity aids in the control of the intermediate grain size in that much broader intermediate annealing temperature ranges may be employed. While sulfur and manganese are effective to produce these desired results, other and specifically different elements are capable of being substituted for manganese. For example, an alloy having a nominal 3.3 percent silicon content, 0.09 percent titanium, and 0.032 percent sulfur, balance substantially all electrolytic iron to which no manganese was added, and a substantially identical alloy containing 0.13 percent chromium instead of the titanium, and 0.033 percent sulfur, were prepared by the procedure disclosed for alloys 1-7. It was found that these alloys exhibited secondary recrystallization up to a final annealing temperature of about 1175° C. for the titanium alloy, and up to a final annealing temperature of about 1075° C. for the chromium alloy, providing the intermediate annealing temperature did not exceed 1000° C. and the intermediate grain size lay between about 0.010 and 0.030 mm.

In Fig. 2 is plotted the effect of impurity concen-

tration, expressed as percent sulfur, upon the intermediate grain size obtained by means of a 900° C. anneal for 15 minutes. It will be understood that the alloys contain a sufficient amount of another material such as manganese, titanium, or chromium, for example, to combine with sulfur to form their respective sulfides. It will be seen that such alloys should contain at least about 0.015 percent sulfur in order that an intermediate grain size between 0.010 and 0.030 mm. may be readily achieved and which develop secondary recrystallized grain structure during the final annealing treatment.

It will, of course, be appreciated that currently the commercially produced alloys are made by air melting practices using relatively impure materials and hence have many undesirable materials present as second phase inclusions such as sulfides, oxides and nitrides, for example, in varying degrees of size, distribution and having varying degrees of stability. As is well known, many of these impurities have very substantial deleterious effects upon the magnetic properties of the finished strip or sheet material and, in order to eliminate these small amounts of undesirable impurities with certainty, vacuum melting and casting techniques utilizing high purity materials such as previously described are necessary. It will thus be appreciated that producers of these high purity materials must be aware of the various processing and compositional variables which I have disclosed in order that the highest quality material may be consistently produced; however, it will be apparent that these processing and compositional variables are equally applicable to commercially produced materials. Further, in view of the rate at which secondary recrystallization may be accomplished in thin strip or sheet materials according to my invention, it will be apparent to those skilled in the art that such materials resulting from the practice of my invention may be successfully processed by means of continuous annealing techniques rather than the time consuming batch annealing process.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. The method of fabricating polycrystalline sheet-like bodies of metal comprising electrical grade silicon-iron alloy consisting essentially of from about 2.5 to 4.0 percent silicon, more than about 0.012 percent sulfur, a sufficient amount of an element selected from the group consisting of manganese, titanium and chromium to combine with the sulfur to form sulfide and the balance substantially all iron comprising the steps of cold reducing an at least partially recrystallized body of said alloy at least 40 percent to form a body of intermediate thickness, heat treating said cold reduced body at a temperature of between 700° C. to 1000° C. to produce a measured average grain size of from about 0.010 mm. to about 0.030 mm., cold reducing said annealed body at least 40 percent by unidirectional rolling to produce a sheet-like body of final thickness and raising the temperature of said cold worked sheet-like body to a temperature of at least 900° C. for a time sufficient to effect secondary recrystallization of the material.

2. The process as set forth in claim 1 in which said sulfur content is at least 0.018 percent and said intermediate thickness worked body is annealed at a temperature of about 700° to 950° C. to produce an average measured grain size of from about 0.010 to about 0.025 mm.

3. The process as set forth in claim 1 in which the alloy contains at least about 0.025 percent manganese.

4. The process as set forth in claim 1 in which the alloy contains at least about 0.025 percent titanium.

5. The process as set forth in claim 1 in which the alloy contains at least about 0.025 percent chromium.

References Cited in the file of this patent

UNITED STATES PATENTS

2,158,065 Cole et al. May 16, 1939