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3,090,711

PROCEDURE FOR SECONDARY RECRYSTALLIZATION

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This invention relates to the manufacture of silicon-iron sheet stock having a high degree of preferred orientation primarily produced by secondary recrystallization from a suitable starting material. The invention will be described in connection with the manufacture of silicon-iron sheet stock characterized preponderantly by a (100)[001] crystal orientation, hereinafter referred to as a "cubic texture," but is not necessarily so limited, as will hereinafter be pointed out.

It has been understood in the art that a silicon-iron sheet stock having a high degree of cubic texture would be a useful material in the electrical arts. It would be a material in which the cube faces of the crystals lie parallel to or within a few degrees of parallelism to the surface planes of the sheet stock, while the cube edges are generally aligned with the rolling direction. As a consequence, such a material could be expected to have, and does in fact have, as high a permeability (or higher) in the straight grain or rolling direction as characterizes silicon-iron having the so-called cube-on-edge orientation, but also a high permeability in the transverse direction. Hence it would be especially useful in the manufacture of transformers using core stampings, and in rotating electrical machinery.

While a number of ways of making a silicon-iron sheet stock having a cubic texture have hitherto been suggested in the art, the only processes which have commercial economy are those in which a material having, as a result of previous treatments and a final primary recrystallization, a reasonable number of grains oriented in the (100)[001] direction or near it, is subjected to a high temperature secondary recrystallization during which these grains grow at the expense of grains having a substantially different orientation, until the sheet attains a condition in which the cubic texture predominates.

Various ways have been suggested for producing a material of desired gauge which is characterized by a reasonable number of cubic nuclei. In a copending application of the inventor and Martin F. Littmann, entitled Oriented Silicon-Iron and Process of Making It, Serial No. 816,889, filed May 29, 1959, there is described a procedure in which the starting material is commercially oriented silicon-iron having at (110)[001] or "cube-on-edge" crystal orientation by Miller's indices. This starting material, by a series of cold rolling treatments and intermediate and final primary recrystallizations is carried through a series of well defined derivative orientations to a condition in which it has a relatively large number of crystal nuclei in the cubic orientation, whereupon it is subjected to secondary recrystallization. In another copending application entitled The Manufacture of Silicon-Iron Having Cubic Texture, Serial No. 819,589, filed June 11, 1959, the same inventors have described a process by which a material having a satisfactory number of cubic crystal nuclei may be produced from hot rolled strip stock in a series of two cold rollings and intermediate and final primary recrystallization treatments. The starting material for this invention may be made by following steps of either of these processes, or by any other processes which will yield a final gauge product having a reasonable number of cubic crystal nuclei; and the reference to the said copending application is not given for purposes of limitation but rather for the purpose of in-

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corporating herein teachings which will result in the production of the desired starting material. The processes of the said copending applications are preferred because they work very well on air-melted silicon-iron stock (as produced in an open hearth furnace or arc furnace, by way of example), although they may be used in connection with more expensive vacuum-melted stock if desired.

The (100)[001] orientation which has been mentioned above implies not only an alignment of the cube faces in parallelism or substantial parallelism with the surfaces of the sheet stock, but also a general alignment of the cube edges in the direction of rolling. Other stocks can be made in which the cube faces are parallel or substantially parallel to the sheet stock surfaces, but in which the cube edges are not so aligned, but instead are variously directed and may even be random in their relationship to the rolling direction. A stock of this general character is described in the copending application of John M. Jackson, Serial No. 706,091, filed December 30, 1957, and entitled Non-Directional Oriented Silicon-Iron. Such stocks may be made in any suitable way as by the process of the said copending application, by processes involving the starting of columnar grains at the surfaces as when a phase change takes place in the sheet stock at a substantially constant temperature and the like. A product of this character, in which the cube edges have a random orientation will not be characterized by permeabilities which are as high in the straight grain and cross grain directions as a material having the cubic texture which has been described. But, the product will have a substantially equal permeability in all directions including those intermediate the straight grain and cross grain directions. The teachings of this application are applicable to the manufacture by secondary recrystallization of "wagon wheel" and similar orientations from stocks which contain a reasonable number of nuclei (after final primary recrystallization) having their cube faces lying within from 2° to 5° of parallelism with the sheet stock surfaces and irrespective of the orientation of the cube edges. The conditions hereinafter set forth apply to such stocks.

By silicon-iron in this application is meant a material having a silicon content of substantially 2.5 to 4.0%, and of a high degree of purity including low contents of carbon, sulphur, nitrogen, oxide inclusions and the like. Preferred silicon-iron may contain from 2.90 to 3.30 silicon, a carbon content of the melt of not more than about .030%, which will later be reduced to less than about .005%, about .03 to .15% manganese, the remainder being substantially all iron with a total oxide content which should not be more than about .015% at the start of the routing, and which will preferably be reduced to about .005 by the end of the routing.

Secondary recrystallization is a tricky procedure involving much more than a high temperature heat treatment. In a copending application of the present inventor and John M. Jackson, entitled The Production of Oriented Silicon-Iron Sheets by Secondary Recrystallization, Serial No. 813,289, filed May 14, 1959, there is described an annealing treatment in an inert gas such as argon or helium, or in hydrogen, wherein very small quantities of polar compounds such as oxides of carbon or sulphur, or hydrogen sulfide, are entrained in the annealing gases during the period of the secondary recrystallization. It is believed that these polar compounds are absorbed or adsorbed on the surfaces of the crystals in the sheet stock so as to satisfy the unsatisfied positive bonds thereat, the net result of the procedure being a shifting of the energy levels of crystals having various orientations in the sheet stock in such a way that grains having the (100) plane parallel to the sheet surface be-

come the lowest in energy level, so that during the high temperature heat treatment grains so oriented can grow vigorously as respects other grains having different orientations. The use of this annealing procedure is preferred for purposes of this invention; but the invention is not necessarily confined thereto. It is applicable to any high temperature annealing treatment capable of causing grains having the cubic texture to grow at the expense of other grains.

Various other factors, however, influence the occurrence and vigor of the secondary recrystallization phenomenon. It has been understood that the silicon-iron sheet stock itself should have high purity as above set forth and also that its surfaces should be clean, i.e. substantially free from foreign material other than suitable annealing separators, and in particular free from oxide inclusions of substantial or massive character (although a very thin film of iron oxide on the surfaces of the sheet stock may be tolerated). The surfaces of the sheet stock should be as free as possible from any oxide materials which are not reducible in a high temperature heat treatment in hydrogen.

It is a primary object of this invention to provide a procedure and a condition of the material just before it is subjected to secondary recrystallization, both of which greatly facilitate the preferential growth of low-energy oriented nuclei during secondary recrystallization, and in many instances make possible a perfection of the orientation produced by the secondary recrystallization such as could not otherwise be attained.

This and other objects of the invention which will be set forth hereinafter or will be apparent to one skilled in the art upon reading these specifications, are accomplished by that procedure of which an exemplary embodiment will now be described.

In view of the requirements for cleanliness at the surfaces of the stock various treatments have been attempted. These include pickling, electropolishing, and others. The present inventor has hitherto suggested a treatment of the strip in a water solution containing about equal parts of orthophosphoric acid and hydrogen peroxide. Some improvement in the action during secondary recrystallization was noted; but it was not known whether this improvement was due to a cleaning action or to a chemical addition of some kind absorbed or adsorbed upon the surfaces of the sheet stock.

It has now been found that, aside from the nature and purity of the base metal as hereinabove defined, freedom of its surfaces from foreign matter and inclusions especially of materials which are not reducible in hydrogen at high temperatures, and the use of polar compounds in the annealing atmosphere as taught in the copending application last mentioned above, an extremely important single factor in an otherwise suitable heat treatment for secondary recrystallization, is a physical as distinguished from a chemical condition of the surfaces of the sheet stock. This physical condition has been discovered to be one of smoothness as hereinafter defined. It has been found that a physically smooth surface on a cold rolled sheet stock is a very valuable aid to secondary grain growth. The smoothness referred to does not necessarily require that the sheet surfaces be uniplanar. A rather high "peak to valley" variation as determined by a profilometer measurement can be tolerated if the transition from peaks to valleys is gradual and occupies a substantial dimension in the general plane of the sheet surface. On the other hand variations in the sheet surfaces which are sharp or distinctly angular have been shown to be detrimental. A scratch deliberately formed on a surface of the sheet can be shown to impair drastically the grain growth action upon secondary recrystallization.

While it is conceivable that a satisfactory surface condition could be brought about by electropolishing or chemical polishing, this would require a relatively long

continued treatment, and would involve such great expense as to impair the commercial value of the product.

It has been found that the best and least expensive way of securing the optimum surface condition is to cold roll the product with smooth polished rolls. Under these conditions the surfaces of the sheets become smooth as herein defined and acquire a high luster. Under these particular conditions, the peak to valley measurement as determined by a profilometer also becomes an index of the attainment of the desired smoothness, since the manner in which rolls are polished tends to preclude the existence of widely disparate peaks and valleys.

In a series of tests, it was found that the rolling of the material on polished rolls as hereinafter defined produced a far greater improvement in the desired secondary grain growth (as compared with materials rolled on mills having rolls with normal grinding) than did a chemical polishing of the sheets as ordinarily rolled. In other words samples of silicon-iron cold rolled on polished rolls showed much more cubic secondary grain growth in the same final annealing operation than did samples rolled on conventional mill rolls, or conventionally rolled samples after chemical polishing.

The following table shows the results on secondary grain growth of the use of rolls which have had different finishing treatments:

Effect on Cube Grain Growth	Micro Inch Profilometer Measurements	Usual Method Of Achieving This Roll Finish
Excellent.....	2-1	Buffing.
Good.....	1-5	Grinding with cork or shellac bonded abrasive wheel.
Fair.....	5-12	Grinding with commonly used 160 grit grinding wheel.
Poor.....	12-100	Grinding plus light pangborn of roll.

The profilometer measurements given are those characteristic of the finishing treatments. The effect on grain growth in the secondary recrystallization is necessarily given in qualitative terms due to variations in specific samples, especially where the samples are relatively small; but this classification is the result of extended experience; and it is believed that the effect of the rolls on the silicon-iron sheet stock is the factor producing the greatest improvement in the attainment of a high degree of cubic secondary growth, assuming that the secondary recrystallization is otherwise carried on under favorable conditions.

Hitherto the ordinary commercial roll finishes for the rolling of silicon-iron in orientation processes have had profilometer readings varying from about 10 to about 100 microinches.

In the practice of the invention, while all of the cold rolling passes given the material may be carried on with polished rolls, it is not necessary to do this. The cold rolling reduction in stages prior to the final stage may be carried on with rolls having commercial finishes. The final stage of cold rolling, if desired, may employ rolls with commercial finishes for the greater part of the reduction; but the purposes of this invention will be attained if the last part of the final cold rolling is carried on with polished rolls. An excellent effect can be attained in a single final pass through polished rolls providing the reduction in that pass is great enough to produce the desired smooth surface i.e. a reduction of 2% or more.

In general excellent silicon-iron sheet stock having preponderantly the cubic crystal orientation can be produced from a material which in a condition of final primary recrystallization is characterized by an alignment of at least about 70% of the cube edges to within 20° of the rolling direction and an angular relationship of the cube faces of a substantial number of the grains to within at most about 5° of parallelism with the sheet surfaces. The skilled worker in the art will understand that primary

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recrystallization will occur at temperatures roughly between 1400° and 1700° F., and that secondary recrystallization requires a temperature of substantially 1900° to 2300° F. Since the primary recrystallization occurs very rapidly, it is usual for economic reasons to combine a final primary recrystallization with the secondary recrystallization, the primary recrystallization occurring during the heating up of the stock to the high temperatures required for the secondary recrystallization.

Modifications may be made in the invention without departing from the spirit of it. The invention having been described in certain exemplary embodiments, what is claimed as new and desired to be secured by Letter Patent is:

1. In a process of producing silicon-iron sheet stock characterized predominantly by an orientation of the crystals in which the cube faces thereof are substantially parallel to the surfaces of the sheet stock, and including the steps of producing a silicon-iron sheet stock which, after a primary recrystallization will be characterized by a substantial number of grains having their cubic faces so oriented, the production of said stock involving a final cold rolling treatment immediately preceding said primary recrystallization, the improvement which consists in that the said cold rolling treatment is characterized in the final part at least by the reduction of the silicon-iron sheet stock between rolls having a profilometer reading not greater than about 5 microinches before subjecting the stock so produced to a secondary recrystallization treatment at high temperatures.

2. In a process of producing silicon-iron sheet stock characterized predominantly by a cubic orientation of the crystals, and including the steps of producing a silicon-iron sheet stock which, after a primary recrystallization will be characterized by a substantial number of grains having cubic orientation, the production of said stock involving a final cold rolling treatment immediately preceding said primary recrystallization, the improvement which consists in that the said cold rolling treatment is characterized in the final part at least by the reduction of the silicon-iron sheet stock between rolls having a profilometer reading not greater than about 5 microinches

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before subjecting the stock so produced to a secondary recrystallization treatment at a high temperature.

3. The process claimed in claim 2 wherein the said secondary recrystallization is carried on at a temperature substantially between 1900° and 2300° F. in an atmosphere of non-oxidizing gas containing a small quantity of a polar compound selected from a class consisting of oxides of carbon and sulfur and hydrogen sulfide.

4. A process of producing silicon-iron sheet stock having a high degree of cubic texture which comprises subjecting a silicon-iron material containing substantially 2.5 to 4.0% silicon, a carbon content not substantially greater than .005% and an oxide content not substantially greater than .005% to a cold rolling and primary recrystallization treatment in which the crystals thereof are caused to assume an orientation in which at least about 70% of the cube edges are aligned within 20° of the rolling direction, and in which a substantial number of the grains have their cube faces tilted to within substantially 5° of parallelism with the sheet stock surfaces, and thereafter subjecting the stock to a secondary recrystallization, the improvement which consists in accomplishing at least the latter part of said cold rolling by means of polished rolls having a profilometer reading of about .2 to 1 microinch.

5. The procedure claimed in claim 4 wherein said secondary recrystallization is carried on at a temperature of substantially 1900° to 2300° F. in a non-oxidizing atmosphere containing a small amount of a polar compound selected from the group consisting of oxides of carbon and sulfur and hydrogen sulfide.

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