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MAGNETIC STEEL PROCESS

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ABSTRACT OF THE DISCLOSURE

Each face of a sheet or strip of magnetic steel is coated with a thin layer of at least one element of aluminum, silicon, cobalt and germanium, which element is diffused into the sheet until the element is distributed therein in a concentration decreasing from the faces to the core of the sheet, and the resulting product is submitted to heat treatment under the influence of a magnetic field.

This application is a continuation-in-part of Ser. No. 655,005, filed July 21, 1967, and now abandoned.

The present invention relates to a method of improving the magnetic properties of magnetic steels for magnetic applications, and also to the products obtained thereby.

Manufacturers of electrical equipment are becoming more and more particular about the efficiency of electrical machines such as transformers, alternators, etc., and this has led steel specialists to endeavour to develop products in the form of thin sheets for use in construction of magnetic circuits of these machines having constantly improved properties, notably increasingly lower watt losses, since it is the loss of watts in the magnetic circuit that governs the no-load efficiency of a machine and constitutes a constant drawback during the entire useful life of the machine due to the electric power dissipated and lost in the form of heat.

As a rule, the total losses of this character are subdivided into hysteresis losses and eddy-current losses, although such subdivision is disputable from a purely theoretical point of view.

Upon considering the fact that all ferro-magnetic materials consist of juxtaposed elementary Weiss domains, no difference in character should exist between hysteresis losses and eddy-current losses, since both result from induced current at a suitable scale.

To reduce losses, it is particularly advantageous to make these induced currents as low as possible. To this end, two solutions may be contemplated, i.e. either increasing the resistivity or specific resistance of the metal, or in the alternative, modifying in a suitable direction the configuration of the Weiss domains.

The first technique was used from the beginning of this century. Silicon and/or aluminum, or any other suitable elements are added to the iron with a view toward increasing the resistivity of the resulting material.

Another much more recent technique consists of obtaining a metal texture having a predetermined orientation. In properly oriented industrial materials the configuration of the elementary Weiss domains tends to resemble that observed in monocrystals, without in any case attaining their simplicity and perfection.

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In a homogeneous substance having a given resistivity the perfection of the texture orientation constitutes an important factor in that it exerts a direct influence on the configuration of the elementary Weiss domains. It is in this particular manner that it controls all the magnetic properties of the substance, notably the magnitude of its losses.

Japanese Pat. 38/18,068 relates to the manufacture of an aluminum steel by a process which includes deposition of aluminum on iron and a diffusion step. This patent, however, fails to mention the decreasing content of the coating element as required in the present invention. The highest concentration of the element near the surface of the steel sheet brings about a substantial increase in resistivity, precisely in the area where the currents induced in the sheet have the highest value.

Japanese Pat. No. 39/4,697 discloses a magnetic material coated with a thin layer of metal having a greater magnetic permeability than the substrate. However, this process provides a composite product of two materials, i.e. one layer of material bonded to another. The articles produced by the process of this patent fail to provide for the diffusion of the metal into the magnetic material, so as to provide a concentration gradient of the diffused metal.

U.S. Pat. 3,028,269 discloses the coating of certain metals, e.g. zinc and aluminum, on low carbon ferrous metal articles. However, there is no mention therein of the increase in magnetic properties of the coated article, nor of the concentration gradient of the coating metal, i.e. the decrease in concentration of the zinc or aluminum from the surface to the core of the substrate. In fact, it is unlikely that such concentration gradient exists in the articles manufactured by the process of this patent since the temperature of the box anneal following the coating step is too low to cause diffusion of the coating metal. Thus, in the case of zinc, the box anneal is conducted at temperatures up to 550° F. (about 288° C.), and in the case of aluminum, at temperatures up to 650° F. (about 344° C.). These relatively low temperatures (compared to 750–1050° C. in the present invention) would not be sufficient to cause diffusion of the zinc or aluminum into the substrate to cause a concentration gradient, and therefore there would be no significant decrease in the watt losses of the ferrous metal article, as compared to the products produced in the present invention.

U.S. Pat. 2,410,220 is concerned with surface coating, wherein a metal is deposited on a substate and the deposited metal is then oxidized to obtain an oxide which cannot be reduced by hydrogen. Thus, it is apparent that there is no diffusion of the deposited metal into the substrate, and therefore no concentration gradient.

Among the possible orientations two types provide more particularly undeniable technical advantages. The first is the so-called Goss (110) [001] orientation or cube on edge or edged cubical structure, and the second is the so-called cubical (100) [001] orientation or cube laid flat or flat cubical structure. In this case the Millerian indices are used for designating the orientation concerned.

Nowadays metallurgists are capable of manufacturing magnetic materials having these two structure patterns, and the so-called Goss texture made it possible during the last few years to achieve remarkable improvements in the construction of electrical machines in general.

It is an object of the present invention to provide a method whereby substantial improvements in the magnetic properties of steel sheets are obtained, notably in the form of a considerable reduction in the watt losses in relation to those characterizing hitherto known steels for magnetic application.

In fact, this invention permits of obtaining magnetic steel sheets either oriented according to a predetermined texture (such as edged or flat cubical), or non-oriented, containing an additive capable of increasing the resistivity, such as at least one of aluminum, silicon, cobalt and germanium, and any other element capable of exerting a similar effect, at a concentration higher than the values attainable heretofore, with the result that the resistivity of the metal is increased to a degree greatly exceeding that attainable up to the present.

The steel sheets subjected to the process of this invention are already processed, and contain silicon, preferably in an amount of 0.05–6% by weight. However, it is to be understood that the amount of silicon is not critical, and is a function of the presence or absence of other constituent materials in the steel sheet. The English term for these silicon steel sheets is "electrical steel sheets," whereas the French term is "magnetic sheets," these terms bing well-recognized in the art, it being unnecessary to specify the proportion of silicon in the sheets.

As indicated above, the sheets subjected to the process of this invention are already in their processed state. The art is completely unaware of any method whereby the magnetic properties of such processed steel sheets having oriented silicon grains can be substantially improved, by heating such sheets under specific conditions, followed by deposition of the coating element on the sheet, and magnetic treatment under equal specific conditions. The present invention provides such as process, wherein the resultant products have vastly superior magnetic properties compared to those previously known.

The present invention is characterized mainly by the fact that it comprises coating each face of a magnetic steel product occurring generally in sheet form, whether oriented or not, with a thin layer of at least one element such as aluminum, silicon, cobalt and germanium, causing the element or elements to be diffused into the sheet while preserving the initial texture thereof until the added element or elements is or are properly distributed therein, at a concentration rate decreasing from the surface to the core of the sheet, this distribution leading to the most marked improvement in the reduction of watt losses, and subsequently applying to the resultant steel sheet a heat treatment under the influence of a magnetic field in order to obtain an improvement in the configuration of the surface Weiss domains by an effect of directional order. The total amount of element coated on the steel sheet is preferably within the range 0.05–3.0% by weight of the sheet.

According to one embodiment of the method of this invention, which provides a pronounced improvement in the magnetic properties of the steel sheet, the diffused element is distributed at a concentration rate decreasing from the surfaces to the core of the sheet, irrespective of the type of element contemplated.

According to another embodiment of the method of this invention, an adequate distribution of the selected diffused element is effected with due regard for the nature of the element and the steel sheet thickness.

According to a specific arrangement of the method of this invention, each face of the sheet of the magnetic product is coated with a thin layer of the addition element or elements by resorting to known procedure generally specific to the element being deposited, for example from a volatile compound of the element to be deposited, or by evaporating the element in vacuo.

According to another specific arrangement of the method of this invention, the element coated on the magnetic product is diffused into the sheet by preheating the coated sheet under time-temperature conditions which are subordinate to the element to be deposited on the sheet, and also take into account the desired penetration depth of the element and the components of the reducing atmosphere.

According to the form of actuation of this last arrangement of the method of this invention the sheet is heated at a temperature on the order of 750° C. to 1050° C. during a time period up to 100 hours in an atmosphere of dry hydrogen.

During this heating step the element is diffused into the sheet and therefore a beneficial enrichment of the initial magnetic properties of the product is obtained, the enrichment thus obtained causing on the one hand an increase in the specific resistance of the metal and on the other hand a substantial improvement in the efficiency of a treatment of directional order, with the result that watt losses are appreciably reduced in the initial product while preserving the recrystallization characteristics obtained during the normal processing or manufacture of the magnetic product.

It is known the Goss texture is obtained by effecting a secondary recrystallization at a temperature in excess of 1100° C. and that it contains very few pattern defects, whereby a perfect stability is imparted thereto. Then aluminum or one of the other beneficial elements can be diffused according to this invention, while preserving this oriented texture. Only when the heating conditions are maintained within the defined limits is it possible to conserve the edged cubical or flat cubical structure of the steel sheet.

This invention provides two additional and extremely advantageous effects:

(1) For each total concentration of alloy elements there is an optimum distribution of the element added by coating and diffusion. This distribution is not a uniform concentration in the sheet thickness. This invention requires a greater concentration of the element on the sheet faces, this concentration decreasing towards the core of the sheet, since the major induced currents circulate on the faces where the Weiss domains of complex shapes are found, as they are induced by all the defects or imperfections of the orientation and the nature of the crystal surfaces, which domains have been given the name closing domains. For instance, considering strips having a thickness of 0.35 mm. (.01375"), the diffusion treatment giving the optimum conditions is such that the product Dt expressed in C.G.S. units has a value of from 10^{-4} to 10^{-6} sq. cm., wherein D denotes the coefficient of diffusion in square centimeters per second of the element contemplated, at the treatment temperature, and t designates the diffusion treatment time in seconds.

(2) The surface enrichment of the magnetic product permits a modification in the surface or wall energy that can be exploited for producing oriented crystallization of a novel type.

In addition to the arrangements set forth hereinafter, this invention further comprises other arrangements which will readily appear to those conversant with the art from the following more detailed description.

This invention is particularly concerned with a method for improving the magnetic properties of steels for magnetic applications and with the products derived therefrom according to the provisions set forth hereinabove, and also with the elements for carrying out the method and obtaining the products, and the assemblies in which the method is included and the products are used.

This invention will be better understood from the description given hereinafter with reference to a typical specific embodiment of the method constituting the subject matter of this invention, which is given by way of illustration and should not be construed as limiting the scope of the invention.

The initial material is a steel in sheet form prepared according to standard manufacturing specification either with a Goss (110) [001] texture or with a cubical (100) [001] texture, this material having a thickness ranging from .25 to .4 mm. (.01" to 1/64"), and being submitted to the following successive treatments:

The first operation consists in scouring the metal to remove the magnesium silicate layer formed during the final recrystallization annealing. This scouring operation may be carried out in a nitro-hydrofluoric acid bath containing 15% of nitric acid and 4% of hydrofluoric acid, at a temperature of 60° C. After the scouring operation, the metal is washed and dried.

The second operation consists in coating the thus cleaned metal surface with the selected compound, in this instance aluminum. To this end, the metal is treated in a vacuumized enclosure according to the known procedure usually referred to as evaporation in vacuo, and an equal quantity of aluminum is deposited on each face of the steel sheet to be treated. The total quantity to be deposited on the two faces is preferably at the most 3% by weight of the sheet to be treated, and may advantageously approximate 0.5% by weight. In this instance, the total amount is 1.0% by weight.

After the aluminum coating has been applied, the diffusion treatment defined hereinabove is carried out. This treatment consists in effecting a static annealing in a dry hydrogen atmosphere at a temperature of 900° C. during a time period on the order of 3 hours.

Finally, the sheet material is treated in a magnetic field at a temperature of 600° C. to 300° C., preferably in a passage furnace containing a dry hydrogen atmosphere, the intensity of the field being approximately 50 oersteds and its direction being parallel to the preferential direction of easy magnetization of the sheet to be treated. Thus, for example, in the case of sheets having a Goss texture the direction of the magnetic field must be parallel or substantially parallel to the direction in which the sheet was rolled.

This last treatment may be carried out either during the cooling step following the diffusion treatment, or separately, and the cooling rate during this treatment in a magnetic field should range from 5° C. to 300° C. per minute, according to the desired result and the nature of the treated metal.

This treatment in a magnetic field combines very favorably with a hot planishing treatment carried out after the static diffusion annealing. This hot planishing treatment is disclosed in applicants' French Pat. No. 1,426,093 of Nov. 22, 1963.

According to a specific arrangement of the method of this invention, optimum improvement in the magnetic properties of the sheet material is obtained, after the product subjected to the above process has been shaped and just before its actual use, by applying at that time another heat treatment in a passage furnace at a temperature on the order of 750–850° C. during a time period ranging from 1 to 3 minutes, without any protective atmosphere, this heat treatment being followed by cooling from 600 to 300° C. in a magnetic field of about 50 oersteds, the direction of this magnetic field being parallel or substantially parallel to the preferential direction of easy magnetization of the sheet material to be treated.

By applying the method of this invention the watt loss characteristics of the resulting product are improved from .10 to .20 watt per kilogram with a 1.5 Tesla induction and a 50-cycle A.C. frequency.

From the foregoing it is clear that irrespective of the form of embodiment contemplated, the mode of application and the means implemented therefor, there are obtained on the one hand a method capable of bringing substantial improvements in the magnetic properties of steels for magnetic applications, and on the other hand products resulting from the use of such method which, in comparison with hitherto known methods and magnetic products, aiming at the same purpose, provided substantial advantages, notably that of permitting the production of steel

sheets for magnetic applications which have a total content of at least one of aluminum, silicon, cobalt and germanium higher than what has previously been possible, so as to increase very considerably the resistivity of the sheets obtained according to this invention and therefore reduce the watt losses of the sheets and increase the efficiency of a treatment of directional order, without impairing the other properties such as mechanical strength, fragility, etc. of the end product.

Although the present invention has been described in conjunction with a preferred embodiment, it is to be understood that modifications and variations may be resorted to without departing from the spirit and scope of the invention, as those skilled in the art will readily appreciate. Such modifications and variations are considered to be within the purview and scope of the invention and appended claims.

What is claimed is:

1. A process for improving the magnetic properties of processed silicon steel sheets oriented according to a structure belonging to the class including the edged cubical structure or the flat cubical structure, which comprises cleaning the faces of the sheet, coating the cleaned faces of the sheet with a thin layer of at least one element selected from the group consisting of aluminum, silicon, cobalt and germanium, diffusing the element into the sheet while heating the sheet at a temperature of about 750 to 900° C. for a time of up to 100 hours in an atmosphere of dry hydrogen whereby said element is distributed in the sheet in a concentration decreasing from the faces to the core of the sheet, and treating the resultant sheet in a dry hydrogen atmosphere in a magnetic field of approximately 50 oersteds whose direction is substantially parallel to the preferential direction of easy magnetization of the sheet, while cooling the sheet at decreasing temperatures between 600 and 300° C.

2. The process according to claim **1**, wherein subsequent to diffusing the element into the sheet the sheet is submitted to magnetic treatment at a cooling rate of 5 to 300° C. per minute.

3. The process according to claim **2**, wherein the magnetic treatment is carried out in combination with a hot planishing treatment.

4. The process according to claim **1**, wherein the magnetic treatment is carried out in combination with a hot planishing treatment.

5. The process according to claim **1**, wherein the magnetic treatment is followed by a heat treatment at about 750 to 850° C. for a time between 1 and 3 minutes in the absence of a protective atmosphere, said heat treatment then being followed by cooling from 600 to 300° C. in a magnetic field of approximately 50 oersteds whose direction is substantially parallel to the direction of easy magnetization of the sheet.

6. The process according to claim **1**, wherein the coating of each face of the sheet with the element is carried out employing a volatile compound of the element.

7. The process according to claim **1**, wherein the coating of each face of the sheet with the element is carried out by evaporating the element in vacuo.

8. The process according to claim **1**, wherein the element is aluminum and the total quantity of the element coated on the sheet is 0.5 to 3% by weight of the sheet.

9. The process according to claim **1**, wherein the element is cobalt and the total quantity of the element coated on the sheet is 0.05 to 3% by weight of the sheet.

10. The process according to claim **1**, wherein the sheet to be coated has a thickness of about 0.35 mm. and the diffusion treatment is such that the product Dt, in C.G.S. units, is between 10^{-4} and 10^{-6} cm.^{2}, where D is the diffusion coefficient, in cm.^{2}/sec., of the element at the treatment temperature, and t is the time of the diffusion treatment in seconds.

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