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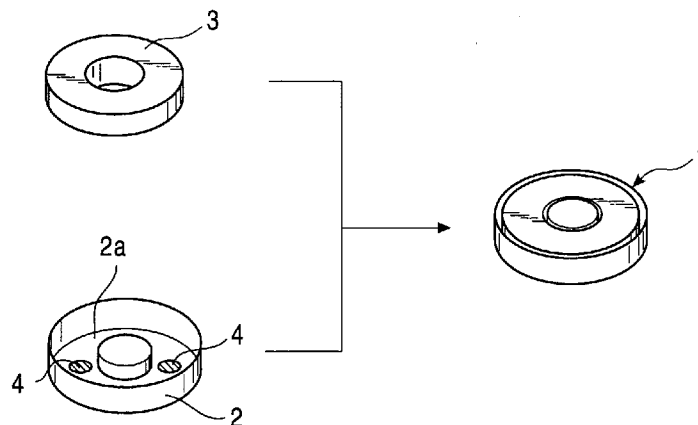
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Amended claims in accordance with Rule 86 (2)
EPC.

(54) Magnetic cores of bulky and laminated types

(57) A bulky magnetic core is disclosed which is composed of a magnetic core body derived by sintering a powdered stock of a soft magnetic glassy alloy, or by cooling a hot melt of the glassy alloy in solidified form, the glassy alloy containing one or more elements selected from Fe, Co and Ni as main components, one or more elements selected from Zr, Nb, Ta, Hf, Mo, Ti and V, and

also B, the glassy alloy having a temperature interval ΔT_x of higher than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature. A laminated magnetic core is also disclosed in which a magnetic core body is derived by tholoidally winding or laminating a ribbon of the above soft magnetic glassy alloy.

FIG. 1**EP 0 899 753 A1**

Description

[0001] This invention relates to a bulky magnetic core and a laminated magnetic core which are composed of a soft magnetic glassy alloy and are suited for use in transformers, choke coils, magnetic sensors and the like.

[0002] A magnetic core of a 50% Ni-Fe permalloy, a magnetic core of a 80% Ni-Fe permalloy, and a silicon steel have heretofore been employed as magnetic core materials for transformers, choke coils, magnetic sensors and the like. However, those magnetic cores resulting from these magnetic materials pose the problem that they cause a great core loss particularly in a high-frequency region and a sharp rise in temperature at a frequency of several tens of kHz or more. Such magnetic cores are generally inapplicable in that frequency region.

[0003] To cope with the problem mentioned above, a certain laminated magnetic core has recently been employed which is constructed with a magnetic core body obtained by tholoidally winding a Co-based amorphous alloy ribbon having a small core loss and a high angular ratio, or a Fe-based amorphous alloy ribbon having a high saturation magnetic flux density and a high maximum magnetic permeation, or by punching such a ribbon into a given shape and then laminating the resultant shapes. During winding or lamination of the ribbon, however, a gapping of 3 μm or so is liable to occur between two adjoining ribbons since the ribbon is or concave and convex on both sides.

[0004] The volume occupied by an alloy ribbon with respect to the volume of a magnetic core body is called a lamination factor. In the above noted instance, the lamination factor is computed as follows:

$$20 \text{ } (\mu\text{m}) / (20 + 3(\mu\text{m})) \times 100 = 87\%$$

This equation shows that the volume of a gapping is large relative to a magnetic core body, meaning that a downsized magnetic core is unfeasible.

[0005] Consequently, a magnetic core derived by laminating an amorphous alloy ribbon leaves the problem that it suffers from a rise in the magnetic flux leaked in between two ribbons, eventually bringing about increased core loss.

[0006] In addition, one technology has been developed in which a powdered stock obtained by grinding the above alloy ribbon is subjected to sintering, followed by solidification of the sintered material in bulky form. This technology requires for the powdered stock to be sintered at a relatively low temperature so as to prevent the same from becoming crystallized, thus failing to produce a magnetic core with high density. Namely, the finished magnetic core is conducive to increased core loss.

[0007] In order to eliminate the foregoing problems of the prior art, the present invention provides a bulky mag-

netic core which has minimized core loss. The invention further provides a laminated magnetic core which has minimized core loss and enables downsizing.

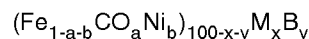
[0008] In accordance with a first aspect of the invention, there is provided a bulky magnetic core comprising a magnetic core body derived by sintering a powdered stock of a soft magnetic glassy alloy, the glassy alloy comprising one or more elements selected from the group consisting of Fe, Co and Ni as main components, one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and B, the glassy alloy having a temperature interval ΔT_x of higher than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature.

[0009] In the bulky magnetic core according to the invention of the first aspect, the magnetic core body may be derived by sintering the powdered stock of the glassy alloy by means of spark plasma sintering and at a speed of temperature rise of 10°C/min or higher.

[0010] In the above composition of the soft magnetic glassy alloy, Zr or Hf may be necessarily present, and the ΔT_x may be higher than 25°C.

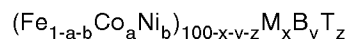
[0011] In the bulky magnetic core according to the invention of the first aspect, the magnetic core body may be derived by cooling a hot melt of the soft magnetic glassy alloy in solidified form.

[0012] In the bulky magnetic core according to the invention of the first aspect, the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and have a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

[0013] In the bulky magnetic core according to the invention of the first aspect, the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and have a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

[0014] More desirably, in the invention of the first aspect, the ΔT_x may be set at higher than 60°C, and the ratio a referred to as a ratio of composition of Co may be set to be $0.042 \leq a \leq 0.29$ and the ratio b referred to

as a ratio of composition of Ni to be $0.042 \leq b \leq 0.43$.

[0015] In the bulky magnetic core according to the invention of the first aspect, the element M may be represented by the formula $(M'_{1-c}M''_c)$ where M' is either one or both of Zr and Hf, M'' is one or more elements selected from the group consisting of Nb, Ta, Mo, Ti and V, and the ratio c is $0 \leq c \leq 0.6$.

[0016] In the above composition of the soft magnetic glassy alloy, the ratio c may be $0.2 \leq c \leq 0.4$ or $0 \leq c \leq 0.2$.

[0017] In the bulky magnetic core according to the invention of the first aspect, the magnetic core body may be derived by heat-treating the soft magnetic glassy alloy at from 427 to 627°C.

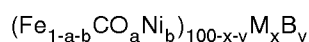
[0018] In the above composition of the soft magnetic glassy alloy, the element B may be replaced in an amount of not more than 50% by an element C.

[0019] In accordance with a second aspect of the invention, there is provided a laminated magnetic core comprising a magnetic core body derived from a ribbon of a soft magnetic glassy alloy, the glassy alloy comprising one or more elements selected from the group consisting of Fe, Co and Ni as main components, one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and B, the glassy alloy having a temperature interval ΔT_x of higher than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature.

[0020] In the above composition of the soft magnetic glassy alloy, Zr may be necessarily present, and the ΔT_x may be higher than 25°C.

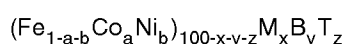
[0021] In the laminated magnetic core according to the invention of the second aspect, the magnetic core body may be derived by tholoidally winding or laminating the ribbon of the soft magnetic glassy alloy.

[0022] In the laminated magnetic core according to the invention of the second aspect, the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and have a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

[0023] In the laminated magnetic core according to the invention of the second aspect, the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and have a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group con-

sisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

[0024] More desirably, in the invention of the second aspect, the ΔT_x may be higher than 60°C, the ratio a referred to as a ratio of composition of Co may be $0.042 \leq a \leq 0.29$, and the ratio b referred to as a ratio of composition of Ni may be $0.042 \leq b \leq 0.43$.

[0025] In the laminated magnetic core according to the invention of the second aspect, the element M may be represented by the formula $(M'_{1-c}M''_c)$ where M' is either one or both of Zr and Hf, M'' is one or more elements selected from the group consisting of Nb, Ta, Mo, Ti and V, and the ratio c is $0 \leq c \leq 0.6$.

[0026] In the above composition of the soft magnetic glassy alloy according to the invention of the second aspect, the ratio c may be $0.2 \leq c \leq 0.4$ or $0 \leq c \leq 0.2$.

[0027] In the laminated magnetic core according to the invention of the second aspect, the magnetic core body may be derived by heat-treating the soft magnetic glassy alloy at from 427 to 627°C.

[0028] In the above composition of the soft magnetic glassy alloy according to the invention of the second aspect, the element B may be replaced in an amount of not more than 50% by C.

BRIEF DESCRIPTION OF THE DRAWINGS

[0029] FIG. 1 is an exploded view of the bulky magnetic core according to the present invention.

[0030] FIG. 2 is a cross-sectional view showing important parts of one form of a spark plasma sintering apparatus for use in producing the bulky magnetic core of the invention.

[0031] FIG. 3 is a view showing one form of a pulse current wave applied to a powdered stock by the spark plasma sintering apparatus.

[0032] FIG. 4 is an exploded view of the laminated magnetic core according to the invention.

[0033] FIG. 5 is an exploded view showing a modified form of the laminated magnetic core according to the invention.

[0034] FIG. 6 is a graphic representation as to the DSC curves of those ribbon specimens of the glassy alloys composed of $Fe_{60}Co_3Ni_7Zr_{10}B_{20}$, $Fe_{56}Co_7Ni_7Zr_{10}B_{20}$, $Fe_{49}Co_{14}Ni_7Zr_{10}B_{20}$ and $Fe_{46}Co_{17}Ni_7Zr_{10}B_{20}$, respectively.

[0035] FIG. 7 is a triangular diagram as to the dependence of the $\Delta T_x (= T_x - T_g)$ upon the contents of Fe, Co and Ni in regard to a composition of $(Fe_{1-a-b}Co_aNi_b)_{70}Zr_{10}B_{20}$.

[0036] FIG. 8 is a graphic representation as to the X-ray diffraction patterns of quenched ribbons with varying sheet thicknesses in regard to a composition of $Fe_{56}Co_7Ni_7Zr_{10}Nb_6B_{20}$.

[0037] FIG. 9 is a graphic representation as to the dependence of saturation magnetic flux density (B_s), magnetic retentivity (H_c), magnetic permeability (μ_e) at 1

kHz and magnetostriction (λ_s) upon the content of Nb in regard to specimens of compositions of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_{10-x}\text{Nb}_x\text{B}_{20}$ where x is 0, 2, 4, 6, 8 and 10 at. %.

[0038] FIG. 10 is a graphic representation as to the core loss of each of bulky magnetic cores produced from magnetic core bodies of a composition of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{20}$.

[0039] FIG. 11 is a graphic representation of the relationship between the sheet thickness and the lamination factor in regard to the glassy alloy according to the invention.

[0040] FIG. 12 is a graphic representation of the relationship between the core loss and the B_m in regard to each of laminated magnetic cores produced from ribbons of a composition of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{20}$.

[0041] FIG. 13 is a graphic representation of the relationship between the core loss and the B_m in regard to each of laminated magnetic cores produced from ribbons of a composition of $\text{Fe}_{62}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{14}$.

[0042] The bulky magnetic core according to an embodiment of the present invention is now described.

[0043] The bulky magnetic core of the invention is attainable for example in an annular shape. This annular bulky magnetic core may be produced from a magnetic core body which is derived by sintering a powdered stock of a soft magnetic glassy alloy to be described hereinafter, or by casting a hot melt of such glassy alloy in a predetermined mold, followed by cooling of the hot melt in solidified form. The magnetic core body is then covered with for example an epoxy resin, or is encapsulated in a resinous casing for insulating protection.

[0044] In producing a bulky magnetic core for use in an EI core, a magnetic core body is obtained by sintering a powdered stock of a soft magnetic glassy alloy to form an E core and an I core and then by bringing both cores into integrally bonded relation to each other.

[0045] The resultant magnetic core body is covered at a necessary portion thereof with for example an epoxy resin, or is encapsulated in a resinous casing for insulating protection of a necessary portion thereof, whereby the bulky magnetic core for the EI core is provided.

[0046] In FIG. 1, there is shown one preferred embodiment of the annular bulky magnetic core 1 according to the present invention. A bulky magnetic core 1 is constructed with a magnetic core body 3 derived by sintering a powdered stock of a soft magnetic glassy alloy to be described later, or by casting a hot melt of such glassy alloy in a given mold and thereafter by cooling the hot melt in solidified form, and with a casing 2 made of a resin in a hollow annular shape in which the magnetic core body 3 is accommodated.

[0047] The casing 2 may be formed preferably from polyacetal resin, polyethylene terephthalate resin or the like.

[0048] At two separate places on an inner surface of a bottom portion 2a of the casing 2 as viewed in FIG. 1, an adhesive 4 is coated to stably secure the magnetic

core body 3 to the casing 2. Desirably, the number of places to be coated with the adhesive 4 may be in the range of 2 to 4.

[0049] The adhesive 4 is chosen from epoxy resin, silicone rubber and the like.

[0050] Subsequently, a process is described for producing the bulky magnetic core 1 of the present invention by means of spark plasma sintering.

[0051] FIG. 2 shows important parts of one form of a spark plasma sintering apparatus which is suitable for use in the production of the bulky magnetic core 1 according to the invention. This form of spark plasma sintering apparatus is constructed essentially with a cylindrical die 11, an upper punch 12 and a lower punch 13, both punches being inserted in the die 11, a punch electrode 14 disposed to support the lower punch 13 and to act as an electrode on one side in flowing a pulse current to be described later, a punch electrode 15 located to downwardly press the upper punch 12 and to act as an electrode on the other side in flowing the pulse current, and a thermocouple 17 arranged to measure the temperature of a starting powder 16 held in sandwiched relation to the upper and lower punches 12, 13.

[0052] In those faces of the the upper and lower punches 12, 13 opposed to each other, molds are defined which correspond to the shape of a magnetic core body to be formed.

[0053] Additionally, the important parts of the spark plasma sintering apparatus stated above are placed in a chamber not shown. This chamber is connected to a vacuum exhaust system not shown and to an ambient gas supply system not shown such that the starting powder (powdered stock) 16 to be filled in between the upper and lower punches 12, 13 can be maintained in a desirable atmosphere such as in an inert gas atmosphere or the like.

[0054] In order to produce the bulky magnetic core 1 by the use of the spark plasma sintering apparatus constructed above, a powdered stock 16 to be molded is first prepared. The powdered stock 16 may be obtained by melting a soft magnetic glassy alloy of a given composition to be described later and thereafter by subjecting the melt to casting, to quenching with use of a single roll or a twin roll, to solution spinning or solution extraction, or to spraying with use of a high-pressure gas, thereby forming the melt into various shapes including bulky, ribbon-like, linear, powdery and other shapes, and further by granulating the shapes other than a powdery one.

[0055] The soft magnetic glassy alloy for use in the present invention has a temperature interval ΔT_x of higher than 20°C when supercooled to a liquid, or of above 40°C or of above 50°C depending upon the composition of an alloy used. This temperature interval is markedly unique and totally unexpected from those alloys known in the art. Moreover, such glassy alloy is excellent in the soft magnetic properties at room temperature, and hence is unknown and novel.

[0056] Subsequently, the powdered stock 16 prepared above is charged in between the upper and lower punches 12, 13 of the spark plasma sintering apparatus viewed in FIG. 2, and the chamber is drawn into a vacuum in its inside. The powdered stock 16 is molded with pressure applied upwardly downwardly from the two punches 12, 13, and at the same time, it is heated by flow of a pulse current illustrated in FIG. 3 to thereby form a magnetic core body 3 of a desired shape.

[0057] This spark plasma sintering treatment permits the powdered stock 16 to be heated up at a predetermined speed by means of current flow and further enables the temperature of such stock to be strictly controlled according to the value of current flow. Thus, temperature control can be effected with by far greater accuracy than in the case of heating with use of a heater so that sintering is made possible under nearly ideal conditions as designed in advance.

[0058] In the practice of the present invention, it is necessary that the sintering temperature be at 300°C or above so as to mold the powdered stock 16 in solidified form. Since, however, the soft magnetic glassy alloy for use as the powdered stock 16 has a large temperature interval ΔT_x ($T_x - T_g$) when supercooled to a liquid, press sintering in such specific temperature range can preferably produce the magnetic core body 3 having high density.

[0059] However, where the sintering temperature is near to a crystallization temperature, magnetic anisotropy tends to take place due to crystal nucleation (structural ordering in a short range) and to crystallization, making the resultant soft magnetic properties deteriorative.

[0060] Due to the mechanism of the spark plasma sintering apparatus, the sintering temperature to be monitored is a temperature read from the thermocouple 17 attached to the die 11. The temperature so read is lower than that applied to the powdered stock 16.

[0061] For those reasons, the sintering temperature in the invention should be set preferably in the range of $T \leq T_x$ where the crystallization temperature is taken as T_x and the sintering temperature as T .

[0062] According to the present invention, the speed of temperature rise for sintering is preferably higher than 10°C/min. Lower speeds of temperature rise result in the development of undesirable crystalline phases.

[0063] The pressure for sintering is preferably larger than 3 t/cm². Smaller pressures fail to form a magnetic core body.

[0064] The resultant magnetic core body 3 may be heat-treated with the result that its magnetic properties can be enhanced. The temperature for use in the heat treatment is higher than the Curie temperature, but lower than a temperature at which to invite crystals that would be responsible for impaired magnetic properties. More specifically, the heat treatment temperature preferably ranges from 427 to 627°C, more preferably from 477 to 527°C.

[0065] The magnetic core body 3 thus produced has the same composition as that of the soft magnetic glassy alloy used as the powdered stock 16, thus exhibiting superior soft magnetic properties at room temperature. Such magnetic core body when heat-treated is capable of affording its magnetic properties further improved.

[0066] Because of its incorporation with the magnetic core body 3, the bulky magnetic core 1 is excellent in the soft magnetic properties, and hence is widely useful as a magnetic core for transformers, for choke coils and also for magnetic sensors. Hence, magnetic cores are obtainable which are superior in properties to conventional equivalents.

[0067] The foregoing description is directed to the magnetic core body 3 derived by spark plasma-sintering the powdered stock 16 composed of the soft magnetic glassy alloy. Without limitation to this process of sintering, such a magnetic core body may be suitably made obtainable by a sintering process in which pressure is applied as by extrusion.

[0068] In addition, the bulky magnetic core 1 of the present invention can be attained with use of a magnetic core body 3 derived by casting a hot melt of the above soft magnetic glassy alloy in a given mold and then by cooling the melt in solidified form.

[0069] This hot melt may be derived by weighing such starting materials as pure metals of Fe, Co, Ni and Zr, pure crystalline boron and the like in their respective given amounts and then by dissolving those materials in an Ar atmosphere and in vacuo for example by a high-frequency induction heater, an arc furnace, a crucible furnace, a reflective furnace or the like.

[0070] The resulting hot melt of alloy is cast in a mold of a given shape, followed by gradual cooling of the melt in solidified form, whereby a magnetic core body 3 is provided with a desired shape.

[0071] The magnetic core body 3 thus obtained, like that derived by sintering the powdery alloy, is high in its density and excellent in its soft magnetic properties and hence applicable as a magnetic core for use in transformers, choke coils, magnetic sensors and the like.

[0072] Next, the laminated magnetic core according to the present invention is described with reference to the drawings.

[0073] The laminated magnetic core of the invention may be achieved for example in an annular shape. This annularly laminated magnetic core may be produced from a magnetic core body which is derived by forming a ribbon of a soft magnetic glassy alloy to be described later and then by tholoidally winding the ribbon, or by press-punching the ribbon into a plurality of rings and then by laminating the rings in a given number with each other. The resultant magnetic core body is further covered with for example an epoxy resin, or is further encapsulated in a resinous casing for insulating protection.

[0074] In the production of a laminated magnetic core for use in an EI core, a magnetic core body is obtained by press-punching the above soft magnetic glassy alloy

ribbon into a plurality of E type leaves and a plurality of I type leaves, respectively, by laminating the E leaves with each other, or the I leaves with each other, thereby forming an E core and an I core, and subsequently by bringing the E and I cores into integrally bonded relation to each other. The resultant magnetic core is covered at a necessary portion thereof with for example an epoxy resin, or is encapsulated in a resinous casing for insulating protection of a necessary portion thereof, whereby the laminated magnetic core for the EI core is provided.

[0075] In FIG. 4, there is shown one preferred embodiment of the laminated magnetic core of an annular shape according to the present invention. A laminated magnetic core 21 is constructed with a magnetic core body 24 obtained by tholoidally winding a ribbon 23 of a soft magnetic glassy alloy to be described later, and with a casing 22 made of a resin in a hollow annular shape in which the magnetic core body 24 is accommodated.

[0076] Desirably, the casing 22 may be formed for example from polyacetal resin, polyethylene terephthalate resin or the like.

[0077] At two separate places on an inner surface of a bottom portion 22a of the casing 22 as seen in FIG. 4, an adhesive 25 is coated to attach the magnetic core body 24 in a stable posture to the casing 22. The number of places to be coated with the adhesive 25 may be preferably in the range of 2 to 4.

[0078] The adhesive 25 is chosen from epoxy resin, silicone rubber and the like.

[0079] FIG. 5 shows another embodiment of the laminated magnetic core of an annular shape according to the invention. The laminated magnetic core 31 is constructed with a magnetic core body 33 obtained by laminating rings punched out of the ribbon 23 of the soft magnetic glassy alloy to be described later, with a casing 32 made of a resin in a hollow annular shape in which the magnetic core body 33 is accommodated, and also with a cover 34 to be fitted to the casing 32 after the magnetic core body 33 is put into the casing 32. The casing 32 and the cover 34 may be formed preferably from polyacetal resin, polyethylene terephthalate or the like.

[0080] Fe system alloy composed of Fe-P-C system, Fe-P-B system, Fe-Ni-Si system and the like have hitherto been known as exerting glass transition. However, these alloys cannot practically be formed into glassy alloys since they show an extremely small temperature width ΔT_x in their supercooled liquid regions.

[0081] As contrasted, the soft magnetic glassy alloy according to the present invention contains one or more of Fe, Co and Ni as main components and has a unique temperature width ΔT_x of higher than 20°C in its supercooled liquid region represented by the equation of $\Delta T_x = T_x - T_g$ (where T_x is a crystallization temperature, and T_g is a glass transition temperature), or a temperature interval of 25 to 60°C or above depending on the composition of an alloy used. This renders it possible to mold

such glassy alloy by gradual cooling and also to mold the same into a ribbon-like or linear shape with a relatively large sectional thickness.

[0082] To gain increased lamination factor, those leaves of amorphous alloys used for the laminated magnetic cores 21, 31 may be made large in thickness.

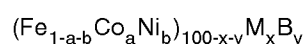
[0083] Conventional amorphous alloys are extremely small in ΔT_x in their supercooled liquid regions as noted above. In the case where a hot melt of a given composition of such an amorphous alloy is rapidly cooled by solution quenching so as to form a ribbon, it is necessary that such ribbon be set to be not more than 50 μm to prevent its magnetic properties from getting diminished. This speaks for a restriction on improved lamination factor.

[0084] The soft magnetic glassy alloy according to the present invention can give a ribbon ranging in sheet thickness from 100 to 200 μm . The magnetic core bodies 23, 33 derivable by winding or laminating such ribbon is so high in lamination factor that downsizing is possible. Further, for its high specific resistance, the ribbon is capable of reducing its core loss as compared in terms of the same sheet thickness to the conventional amorphous alloys.

[0085] The soft magnetic glassy alloy ribbon 23 for use in the laminated magnetic core of the invention may be produced for example by preparing powders of constituting elements, by mixing the powders to meet with in a range of compositions specified above, by dissolving the resulting mixture in an inert gas atmosphere as of an Ar gas and with use of a dissolving device as of a crucible to thereby obtain a hot melt of a selected composition, and subsequently by quenching the hot melt by means of a single roll process. The single roll process noted here denotes a process in which a hot melt is rapidly cooled by being sprayed on a rotating metal roll, whereby a glassy metal is provided with a ribbon-like shape.

[0086] One of the soft magnetic glassy alloys for use in the bulky and laminated magnetic cores described above is composed of one or more selected from the group consisting of Fe, Co and Ni as main components and is further incorporated with one or more selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and B in their given amounts.

[0087] One of the soft magnetic glassy alloys according to the present invention is represented by the following formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ may be preferred, and M is one or more selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

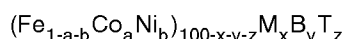
[0088] Besides and importantly, the above composition should have a temperature interval ΔT_x of higher

than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature.

[0089] It is desired, in the above composition, that Zr or Hf be necessarily present and that the ΔT_x be higher than 25°C.

[0090] More preferably, in the above composition, the ΔT_x may be set to be higher than 60°C, the ratio a may be $0.042 \leq a \leq 0.29$, and the ratio b may be $0.042 \leq b \leq 0.43$ may be satisfied in the above defined formula of $(\text{Fe}_{1-a-b}\text{Co}_a\text{Ni}_b)_{100-x-y}\text{M}_x\text{B}_y$.

[0091] Other soft magnetic glassy alloys according to the invention are represented by the following formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P. In the above formula of $(\text{Fe}_{1-a-b}\text{Co}_a\text{Ni}_b)_{100-x-y-z}\text{M}_x\text{B}_y\text{T}_z$, the ratio a may be $0.042 \leq a \leq 0.29$, and the ratio b may be $0.042 \leq b \leq 0.43$ in the invention.

[0092] The above element M may be represented by the formula $(\text{M}'_{1-c}\text{M}''_c)$ where M' is either one or both of Zr and Hf, M'' is one or more selected from the group consisting of Nb, Ta, Mo and V, and the ratio c is $0 \leq c \leq 0.6$.

[0093] The ratio c may further be $0 \leq c \leq 0.4$ or $0 \leq c \leq 0.2$ in the above composition.

[0094] Moreover, the ratio a may be $0.042 \leq a \leq 0.25$, and the ratio b may be $0.042 \leq b \leq 0.1$.

[0095] According to the invention, the soft magnetic glassy alloy may be heat-treated at a temperature of from 427°C (700K) to 627°C (900K). This heat treatment enables a high magnetic permeability to be gained. Additionally, the element B may be replaced in an amount of 50% or below by an element C in the above composition.

[0096] Explanation is provided as to the reasons for the compositions of the soft magnetic glassy alloy to be decided as specified above in implementing the present invention.

[0097] The elements Fe, Co and Ni for use as main components in the invention are necessary for gaining magnetic properties and important for attaining high saturation magnetic flux density and soft magnetic properties.

[0098] More specifically, the ratio a taken as a composition ratio of Co may desirably be set to be $0 \leq a \leq 0.29$ and the ratio b taken as a composition ratio of Ni to be $0 \leq b \leq 0.43$ in order to ensure a ΔT_x of 50 to 60°C. The ratio a taken as a composition ratio of Co may desirably be set to be $0 \leq a \leq 0.29$ and the ratio b taken as

a composition ratio of Ni to be $0 \leq b \leq 0.43$ in ensuring a ΔT_x of 50 to 60°C. To attain a ΔT_x of higher than 60°C, the ratio a may desirably be set at $0.042 \leq a \leq 0.29$ and the ratio b at $0.042 \leq b \leq 0.43$.

[0099] With regard to the above compositions, it is desired that the ratio a taken as a composition ratio of Co be $0.042 \leq a \leq 0.25$ to obtain good soft magnetic properties and that the ratio b taken as a composition ratio of Ni be $0.042 \leq b \leq 0.1$ to gain a high saturation magnetic flux density.

[0100] M is one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V. These elements are effective in rendering the finished alloy amorphous in nature and may be preferably in the range of between above 5 at.% and below 20 at.%. To achieve further enhanced magnetic properties, this range may be between above 5 at.% and below 15 at.%. In particular, Zr is effective among those elements. Part of Zr may be replaced by an element such as Nb or the like, and in such instance, the ratio c may be $0 \leq c \leq 0.6$ so that a high ΔT_x is attainable. To make the ΔT_x of 80°C or higher feasible, the ratio may desirably be $0.2 \leq c \leq 0.4$.

[0101] B has the ability to provide amorphousness, and this element is added in an amount of between above 10 at.% and below 22 at.%. Smaller amounts than 10 at.% lose ΔT_x , failing to produce a magnetic core body 3 of high density, whereas larger amounts than 22 at.% make the resultant magnetic core body brittle. In order to gain improved ability to give an amorphous nature as well as good magnetic properties, the amount of B may be more preferably above 16 at.% but below 20 at.%.

[0102] The compositions specified above may be further incorporated with one or more elements expressed by T and selected from Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

[0103] These elements are added in an amount of between above 0 at.% and below 5 at.%. They are used to principally improve corrosion resistance. Departures from that range are responsible for reduced magnetic properties and also for deteriorated ability to cause amorphousness.

[0104] The soft magnetic glassy alloys of the above specified compositions according to the present invention can offer magnetic properties at room temperature, and upon heat treatment, can produce further improvement in such properties.

[0105] As regards the process for the production of the soft magnetic glassy alloy according to the invention, it should be mentioned that an appropriate cooling speed is decided from the composition of an alloy used, the process means used, the size of a product to be obtained, the shape of a product to be obtained and other parameters. Generally, a range of 10^2 to 10^6 °C/s or so may be taken as a measure of cooling speeds.

[0106] In the bulky magnetic core 1 described hereinbefore, a bulky magnetic core body 3 can be obtained with high density by sintering a powdered stock of a soft

magnetic glassy alloy by means of spark plasma sintering, the soft magnetic glassy alloy having a temperature interval ΔT_x of higher 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature. Hence, reduced core loss is gained.

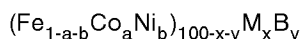
[0107] In addition, in the bulky magnetic core 1, the sintering temperature is optionally selected from a range of temperatures set to meet with the relationship of $T \leq T_x$ where T_x is a crystallization temperature, and T is a sintering temperature. This leads to a magnetic core body 3 that has the same composition as does the soft magnetic glassy alloy for use as a starting material and exhibits high saturation magnetic flux density and excellent magnetic permeability with consequent reduction in core loss.

[0108] Heat treatment of the sintered magnetic core body 3 sintering permits such core body to produce higher saturation magnetic flux density and higher magnetic permeability.

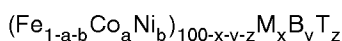
[0109] In the bulky magnetic core 1, the magnetic core body 3 may be formed by a so-called casting process in which a hot melt of an alloy is solidified through cooling, in addition to a spark plasma sintering process. The bulky magnetic core 1 is thus producible with cost savings.

[0110] The above soft magnetic glassy alloy is composed of one or more selected from Fe, Co and Ni as main components, one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V and also of B so that the temperature interval ΔT_x in its supercooled liquid region can be made higher. This means that the powdered stock of the alloy can be sintered at a higher temperature, and the magnetic core body 3 can thus be obtained with higher density. The core loss is therefore small with respect to the bulky magnetic core 1.

[0111] The bulky magnetic core 1 of the invention is provided with a magnetic core body 3 composed of a soft magnetic glassy alloy having a ΔT_x of higher than 50°C, a composition represented by the following formula, high magnetic permeability, low magnetic susceptibility and excellent soft magnetic properties. Reduced core loss is thus made possible.



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V, or

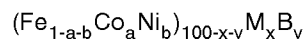


where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more selected from the group consisting of

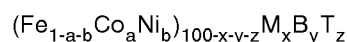
Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more selected from Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

[0112] The laminated magnetic cores 21, 31 described hereinbefore is provided with a magnetic core body 24 derived by tholoidally winding a ribbon of a soft magnetic glassy alloy having a temperature interval ΔT_x of higher 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature, or with a magnetic core body 33 derived by laminating such ribbon. Hence, a magnetic core can be produced from a ribbon of increased sheet thickness so that the lamination factors of the laminated magnetic cores 21, 33 are made large. This ensures reduced core loss and downsized product.

[0113] The above soft magnetic glassy alloy is composed of one or more selected from Fe, Co and Ni as main components, one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V and also of B so that the temperature interval ΔT_x in its supercooled liquid region can be made higher. Accordingly, the laminated magnetic cores 21, 31 can be produced from a ribbon of increased sheet thickness with improved lamination factor and with reduced core loss. The laminated magnetic cores 21, 31 of the invention are provided with magnetic core bodies 24, 33 composed of a soft magnetic glassy alloy having a ΔT_x of higher than 50°C, a composition represented by the following formula, high magnetic permeability, low magnetic susceptibility, high saturation magnetic flux density and excellent soft magnetic properties. Small core loss is thus attainable.



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V, or



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more selected from Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more selected from Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

EXAMPLES

Example 1

[0114] Pure metals of Fe, Co, Ni and Zr, and pure crystalline boron were mixed in an Ar gas atmosphere and melted by arc, whereby a matrix alloy was prepared.

[0115] Subsequently, this matrix alloy was melted with use of a quartz nozzle and subjected to a single roll proc-

ess in which the melt was quenched by jetting on to a copper roll being rotated at 40 m/s in an Ar gas atmosphere from a hole of 0.4 mm in diameter defined at a lower end of the nozzle and at a jet pressure of 0.39×10^5 Pa. There were thus produced ribbon specimens of a glassy alloy which specimens were from 0.4 to 1 mm in width and from 13 to 22 μm in thickness. The resultant specimens were analyzed by differential scanning calorimetry (DSC).

[0116] FIG. 6 shows the DSC curves of those ribbon specimens of glassy alloys composed of $\text{Fe}_{60}\text{Co}_3\text{Ni}_7\text{Zr}_{10}\text{B}_{20}$, $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_{10}\text{B}_{20}$, $\text{Fe}_{49}\text{Co}_{14}\text{Ni}_7\text{Zr}_{10}\text{B}_{20}$ and $\text{Fe}_{46}\text{Co}_{17}\text{Ni}_7\text{Zr}_{10}\text{B}_{20}$, respectively.

[0117] Each of the specimens has been found to have a wide region of a supercooled liquid as the temperature increased and to get crystallized when the temperature exceeded the supercooled liquid region. The temperature interval ΔT_x in the supercooled liquid region is represented by the equation $T_x = T_x - T_g$, and the value of $T_x - T_g$ of each specimen is beyond 60°C and in the range of 64 to 68°C as is clear from FIG. 6. A substantial equilibrium state indicative of a supercooled liquid region was obtained in a wide range of from 596°C (869K) slightly lower than a temperature indicative of crystallization due to an exothermic peak to 632°C (905K).

[0118] FIG. 7 is a triangular diagram as to the dependence of the $\Delta T_x (= T_x - T_g)$ upon the contents of Fe, Co and Ni in regard to a composition of $(\text{Fe Co Ni})_{70}\text{Zr}_{10}\text{B}_{20}$.

[0119] As is evident from FIG. 7, the value of ΔT_x is beyond 25°C in all ranges of the composition of $(\text{Fe Co Ni})_{70}\text{Zr}_{10}\text{B}_{20}$.

[0120] With regard to the value of T_g , it has been found that T_g is monotonously increased when Co is increased in a range of about 7 at. % to about 50 at. %. On the other hand, with regard to ΔT_x , it has also been found that the value of ΔT_x is large in a Fe-abundant composition as is apparent from FIG. 7 and that in order to gain a ΔT_x of higher than 60°C , the content of Co is preferably in the range between above 3 at. % and below 20 at. %, and the content of Ni is preferably in the range between above 3 at. % and below 30 at. %.

[0121] The above composition is expressed in terms of the ratios Fe, Co and Ni as $(\text{Fe}_{1-a-b}\text{Co}_a\text{Ni}_b)_{70}\text{Zr}_{10}\text{B}_{20}$. Thus, for Co to be more than 3 at. % in content the ratio a of Co is above 0.042, and for Co to be less than 20 at. %, the ratio a is below 0.29 because $(\text{Fe}_{1-a-b}\text{Co}_a\text{Ni}_b)$ is 70 at. %. For Ni to be more than 3 at. % in content, the ratio b of Ni is above 0.042, and for Ni to be less than 30 at. %, the ratio b is below 0.43.

Example 2

[0122] Pure metals of Fe, Co, Ni and Zr, and pure crystalline boron were mixed in an Ar gas atmosphere and melted by arc, whereby a matrix alloy of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_4\text{Nb}_6\text{B}_{20}$ was prepared.

[0123] Subsequently, this matrix alloy was melted

with use of a quartz nozzle and subjected to a single roll process in which the melt was quenched by jetting on to a copper roll being rotated at 40 m/s in an Ar gas atmosphere. There were thus produced ribbon specimens of a glassy alloy.

[0124] Alloy ribbons of 20 to 195 μm in sheet thickness were obtained with suitable adjustments made to the aperture of a nozzle, the distance between a nozzle tip and a roll surface (gap), the revolution of a roll, the jet pressure and the atmospheric pressure.

[0125] X-ray diffraction analysis was made of the specimens with the results shown in FIG. 8.

[0126] As evidenced by this figure, each of the specimens has been found to have a hollow pattern at $2\theta = 38$ to 52° and hence an amorphous structure of a single layer.

Example 3

[0127] The procedure of Example 1 was repeated except that compositions of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_{10-x}\text{Nb}_x\text{B}_{20}$ where x is 0, 2, 4, 6, 8 and 10 at. % were used. Specimens of glassy alloy ribbons were thus obtained.

[0128] The resultant specimens were then heat-treated at 527°C (800K) for 5 minutes.

[0129] FIG. 9 shows the dependence of saturation magnetic flux density (B_s), coercive force (H_c), Magnetic permeability (μ_e) at 1 kHz and magnetostriction (λ_s) upon the content of Nb in regard to the specimens.

[0130] The saturation magnetic flux density (B_s) decreased with addition of Nb both in a rapidly cooled specimen and a heat-treated specimen. A Nb-free specimen revealed more than 0.9 (T), while a specimen containing 2 at. % of Nb revealed about 0.75 (T).

[0131] The magnetic permeability (μ_e) was 5,031 in a specimen quenched but containing no Nb and 2,228 in a specimen quenched and containing 10 at. % of Nb. This permeability (μ_e) fell at 906 in a quenched specimen containing 10 at. % of Nb. Heat treatment, however, led to sharply improved permeability (μ_e) and showed about 25,000 particularly in a specimen containing 2 at. % of Nb.

[0132] The magnetic retentivity (H_c) was as low as 50 A/m ($= 0.625 \text{ Oe}$) both in a Nb-free specimen and a specimen containing 2 at. % of Nb. In particular, a specimen containing less than 2 at. % of Nb showed a remarkably good value, say 5 A/m ($= 0.0625 \text{ Oe}$). Heat treatment ensures excellent magnetic retentivity (H_c) even in a specimen containing more than 4 at. % of Nb.

[0133] It has been found, as evidenced above, that Nb be added in an amount between above 0 at. % and below 2 at. % so as to obtain good soft magnetic properties in the alloy specimens of the foregoing composition. Consequently, a bulky magnetic core or a laminated magnetic core can be produced which is composed of a soft magnetic glassy alloy having high saturation magnetic flux density, low magnetic retentivity and high magnetic permeability. In the case of manufacture of a

transformer with use of such a magnetic core, the finished transformer is made feasible with small core loss and with good power transmission efficiency.

Example 4

[0134] The procedure of Example 1 was repeated except that a composition of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{20}$. Glassy alloy ribbons were thus provided.

[0135] These ribbons were then granulated in the atmosphere by the use of a rotar mill. Among the resultant powders, a powder of 53 to 105 μm in particle diameter was screened and used as a starting powder in subsequent steps.

[0136] About 2 g of the starting powder was filled in a die made of WC with use of a hand press and charged in the die shown in FIG. 2. The powder was pressed by the upper and lower punches in a chamber maintained in an atmosphere of 3×10^{-5} torr and was heated by flow of a pulse wave from a power supply system. As seen in FIG. 3, the pulse wave form was so set that 12 pulses were flowed and 2 pulses interrupted. The powder was heated with a power supply at a maximum of 4,700 to 4,800 A.

[0137] Sintering was carried out by heating the specimen at a pressure of 6.5 t/cm² and from room temperature to a sintering temperature and then by maintaining the same at the latter temperature for about 5 minutes. The speed of temperature rise was 100°C/min.

[0138] A hollow cylindrical specimen of 10 mm in outside diameter, 6 mm in inside diameter and 2 mm in thickness as viewed in FIG. 1 was prepared by spark wire molding, whereby a magnetic core body was obtained.

[0139] This magnetic core body was accommodated in a casing made of a polyacetal resin as shown in FIG. 1. In this instance, a bottom portion of the casing was coated on an inner surface and at two separate places with an adhesive so that the magnetic core body was firmly secured to the casing. Three bulky magnetic cores were produced through the same treatment.

[0140] The core losses of the bulky magnetic cores according to the present invention are shown in FIG. 8.

[0141] In FIG. 10, there is shown the relationship between the working magnetic flux density (B_m) and the core loss with regard to a comparative magnetic core formed by laminating a silicon sheet (Si 3.5%).

[0142] As is apparent from FIG. 10, both the three magnetic cores of the invention and the comparative magnetic core show a rise in core loss as the working magnetic flux densities increase. However, the three magnetic cores provided by the invention are always smaller in core loss than the comparative magnetic core in the range of working magnetic flux densities measured.

Example 5

[0143] In the same manner as in Example 1, glassy alloy ribbons of varying sheet thicknesses were prepared from a composition of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{20}$.

[0144] Subsequently, each ribbon was punched into a ring-like shape, after which the rings were laminated in a given number. epoxy or polyimide resin was immersed in between the layers to provide insulation of each layer and interlaminar bonding. Thus, a laminated magnetic core of an annular shape with 12 mm in outside diameter, 4 mm in inside diameter and 5 mm in thickness was produced as seen in FIG. 5.

[0145] FIG. 11 shows the relationship between the sheet thickness and the lamination factor with regard to the laminated magnetic core obtained above. The lamination factor was determined by cross-sectionally examining the laminated magnetic core by means of a microscope.

[0146] As evidenced by FIG. 11, the lamination factor improves with increases in sheet thickness and becomes almost constant at or above 97% when the sheet thickness exceeds 100 μm . A laminated magnetic core composed of the soft magnetic glassy alloy according to the invention causes no decline in soft magnetic properties as stated above. A laminated magnetic core is thus attainable with small core loss.

[0147] On the other hand, a known amorphous alloy of $\text{Fe}_{78}\text{Si}_9\text{B}_{13}$ gives only a ribbon of 20 μm or so. In such case, the lamination factor is as low as 87%.

[0148] Since the conventional amorphous alloy is small in ΔT_x , it is necessary to form a ribbon in a thickness of not more than 50 μm so as not to impair soft magnetic properties in the production of such ribbon by rapidly cooling a hot melt of an alloy of a given composition through a solution quenching process. Because larger thicknesses than 50 μm result in the reduction of soft magnetic properties, increased lamination factor and improved soft magnetic properties cannot be achieved in a well-balanced manner. This fails to produce a laminated magnetic core of reduced core loss.

Example 6

[0149] In the same manner as in Example 1, a 20 μm thick ribbon of a glassy alloy of $\text{Fe}_{56}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{20}$ and a 20 μm thick ribbon of a glassy alloy of $\text{Fe}_{62}\text{Co}_7\text{Ni}_7\text{Zr}_8\text{Nb}_2\text{B}_{14}$ were obtained. Each such ribbon was punched into a ring-like shape, and the rings were laminated as in Example 5. The resultant laminate was heat-treated at 527°C (800K) for 5 minutes.

[0150] Thereafter, the laminate was impregnated with a polyimide resin as in Example 5, whereby a laminated magnetic core was produced.

[0151] FIG. 12 and FIG. 13 show the relationship between the working magnetic flux density (B_m) and the core loss with respect to the laminated magnetic cores obtained above.

[0152] For comparative purposes, the core loss of a laminated magnetic core derived from a silicon steel (Si 6.5%) is shown also in both figures.

[0153] In FIG. 12 and FIG. 13, the results of three different specimens formed by the same treatment are shown.

[0154] The laminated magnetic cores of the invention have proved, as is clear from FIG. 12 and FIG. 13, to be smaller in core loss than the comparative magnetic core.

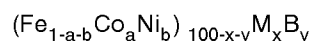
Claims

1. A bulky magnetic core comprising a magnetic core body derived by sintering a powdered stock of a soft magnetic glassy alloy, the glassy alloy comprising one or more elements selected from the group consisting of Fe, Co and Ni as main components, one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and B, the glassy alloy having a temperature interval ΔT_x of higher than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature.

2. A bulky magnetic core according to claim 1, wherein the magnetic core body is derived by sintering the powdered stock of the glassy alloy by means of spark plasma sintering and at a speed of temperature rise of 10°C/min or higher.

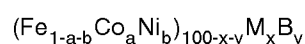
3. A bulky magnetic core according to claim 1, wherein the magnetic core body is derived by cooling a hot melt of the soft magnetic glassy alloy in solidified form.

4. A bulky magnetic core according to claim 1, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



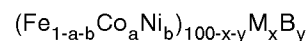
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

5. A bulky magnetic core according to claim 2, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



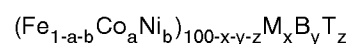
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

6. A bulky magnetic core according to claim 3, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



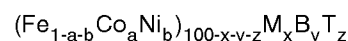
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

7. A bulky magnetic core according to claim 1, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



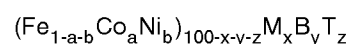
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

8. A bulky magnetic core according to claim 2, wherein the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and has a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

9. A bulky magnetic core according to claim 3, wherein the soft magnetic glassy alloy may have a ΔT_x of higher than 50°C and has a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

10. A bulky magnetic core according to claim 5, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$.

11. A bulky magnetic core according to claim 6, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$.

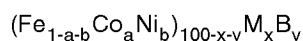
12. A bulky magnetic core according to claim 7, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$.

13. A bulky magnetic core according to claim 8, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$.

14. A laminated magnetic core comprising a magnetic core body derived from a ribbon of a soft magnetic glassy alloy, the glassy alloy comprising one or more elements selected from the group consisting of Fe, Co and Ni as main components, one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and B, the glassy alloy having a temperature interval ΔT_x of higher than 20°C in its supercooled liquid region, the ΔT_x being represented by the equation $\Delta T_x = T_x - T_g$ where T_x is a crystallization temperature, and T_g is a glass transition temperature.

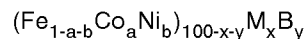
15. A laminated magnetic core according to claim 14, wherein the magnetic core body is derived by tholoidally winding or laminating the ribbon of the soft magnetic glassy alloy.

16. A laminated magnetic core according to claim 14, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 60°C and has a composition represented by the formula



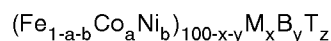
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

17. A laminated magnetic core according to claim 15, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 60°C and has a composition represented by the formula



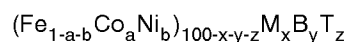
where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$ and $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ are met, and M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V.

18. A laminated magnetic core according to claim 14, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 60°C and the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

19. A laminated magnetic core according to claim 15, wherein the soft magnetic glassy alloy has a ΔT_x of higher than 60°C and the soft magnetic glassy alloy has a ΔT_x of higher than 50°C and has a composition represented by the formula



where $0 \leq a \leq 0.29$, $0 \leq b \leq 0.43$, $5 \text{ at.}\% \leq x \leq 20 \text{ at.}\%$, $10 \text{ at.}\% \leq y \leq 22 \text{ at.}\%$ and $0 \text{ at.}\% \leq z \leq 5 \text{ at.}\%$ are met, M is one or more elements selected from the group consisting of Zr, Nb, Ta, Hf, Mo, Ti and V, and T is one or more elements selected from the group consisting of Cr, W, Ru, Rh, Pd, Os, Ir, Pt, Al, Si, Ge, C and P.

20. A laminated magnetic core according to claim 16, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$.

21. A laminated magnetic core according to claim 17, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$. 5
22. A laminated magnetic core according to claim 18, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$. 10
23. A laminated magnetic core according to claim 19, wherein the ΔT_x is higher than 60°C , the value of a taken as a composition ratio of Co is $0.042 \leq a \leq 0.29$, and the value of b taken as a composition ratio of Ni is $0.042 \leq b \leq 0.43$. 15

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FIG. 1

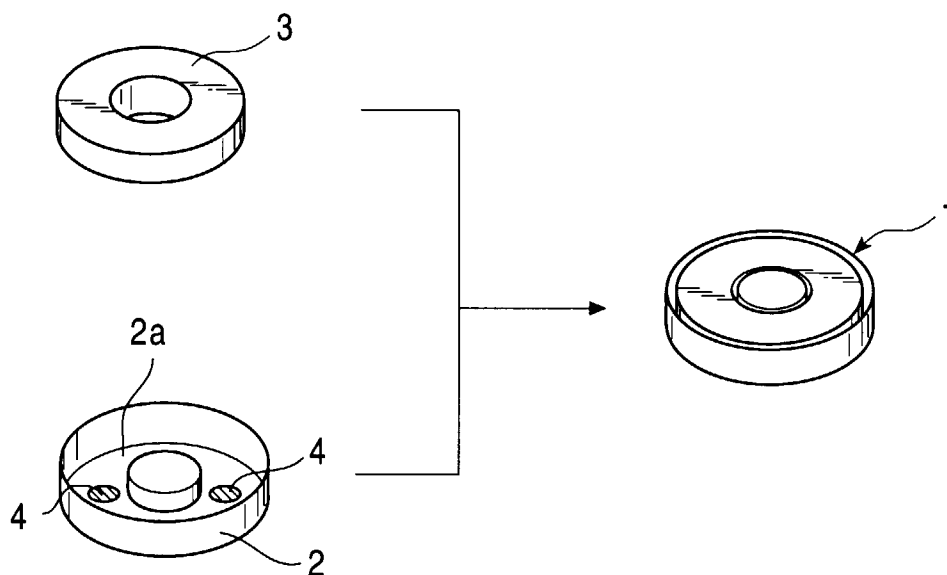


FIG. 2

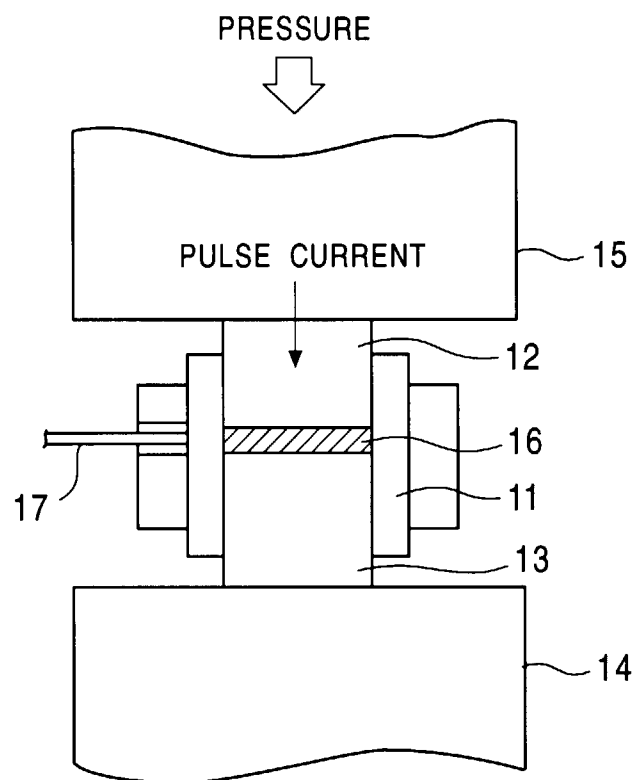


FIG. 3

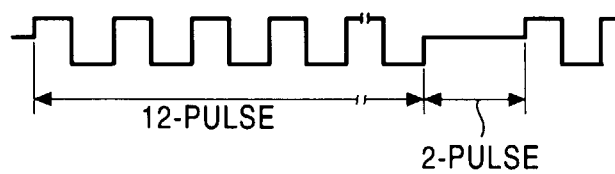


FIG. 4

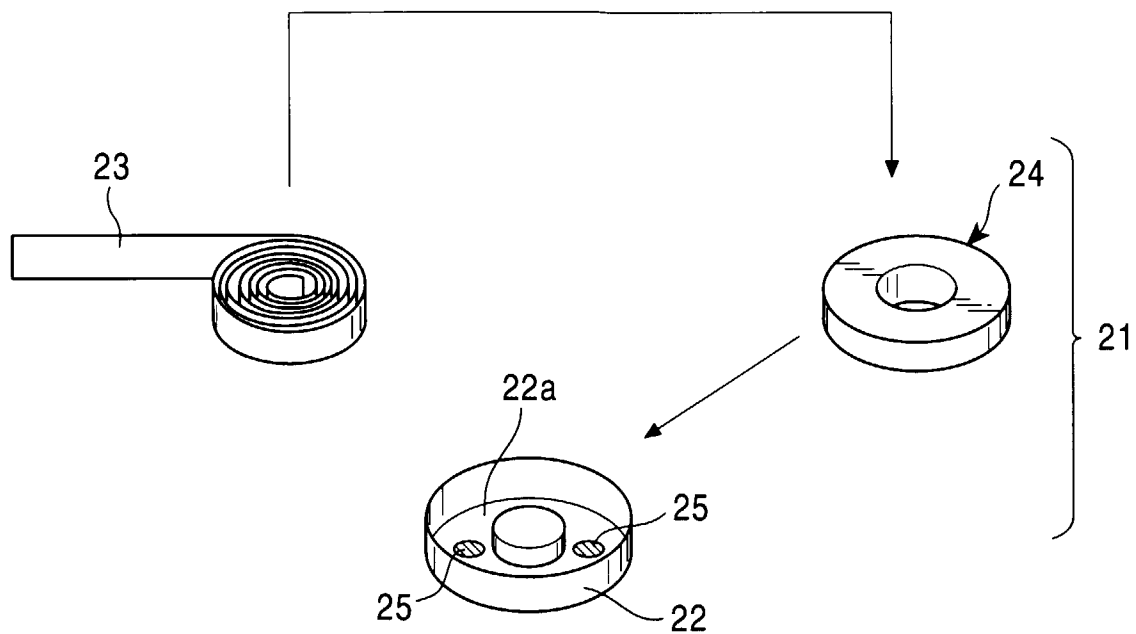


FIG. 5

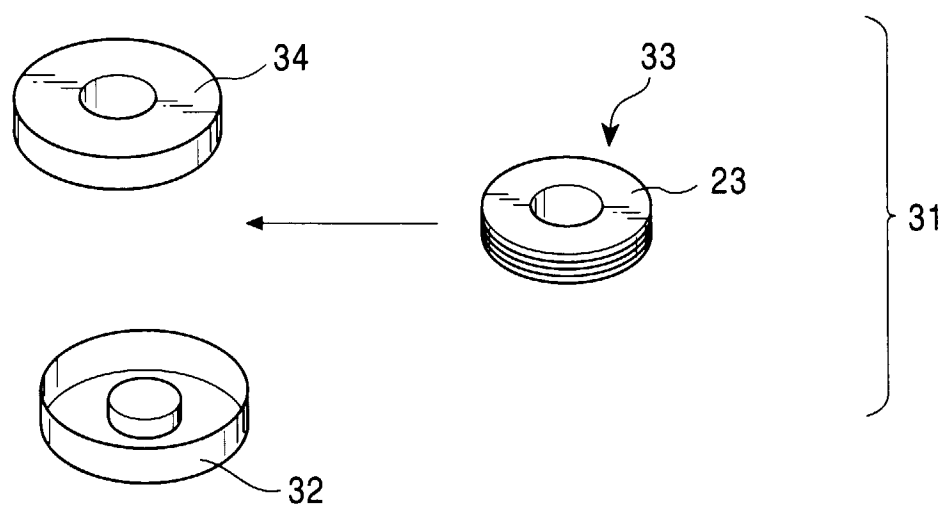


FIG. 6

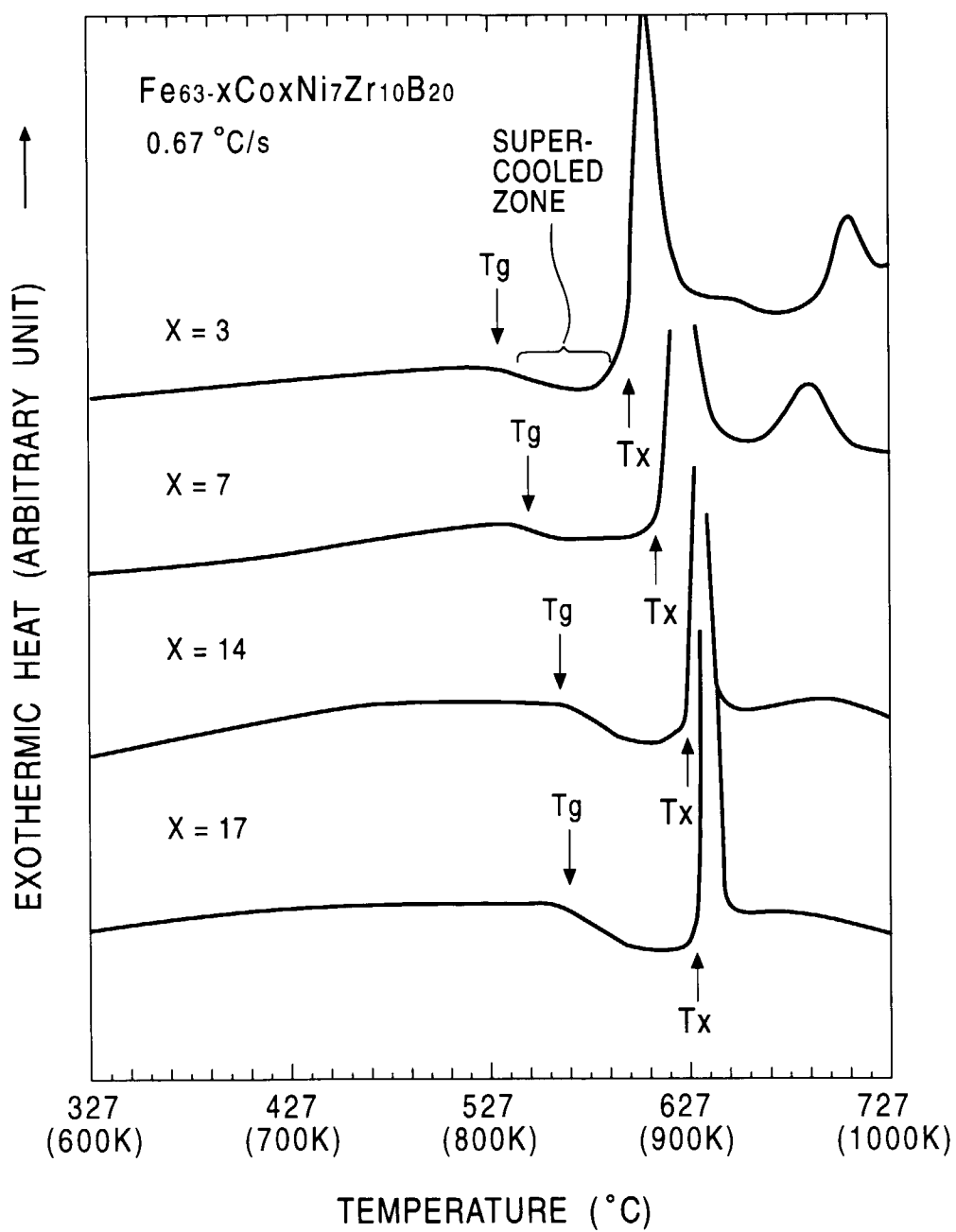


FIG. 7

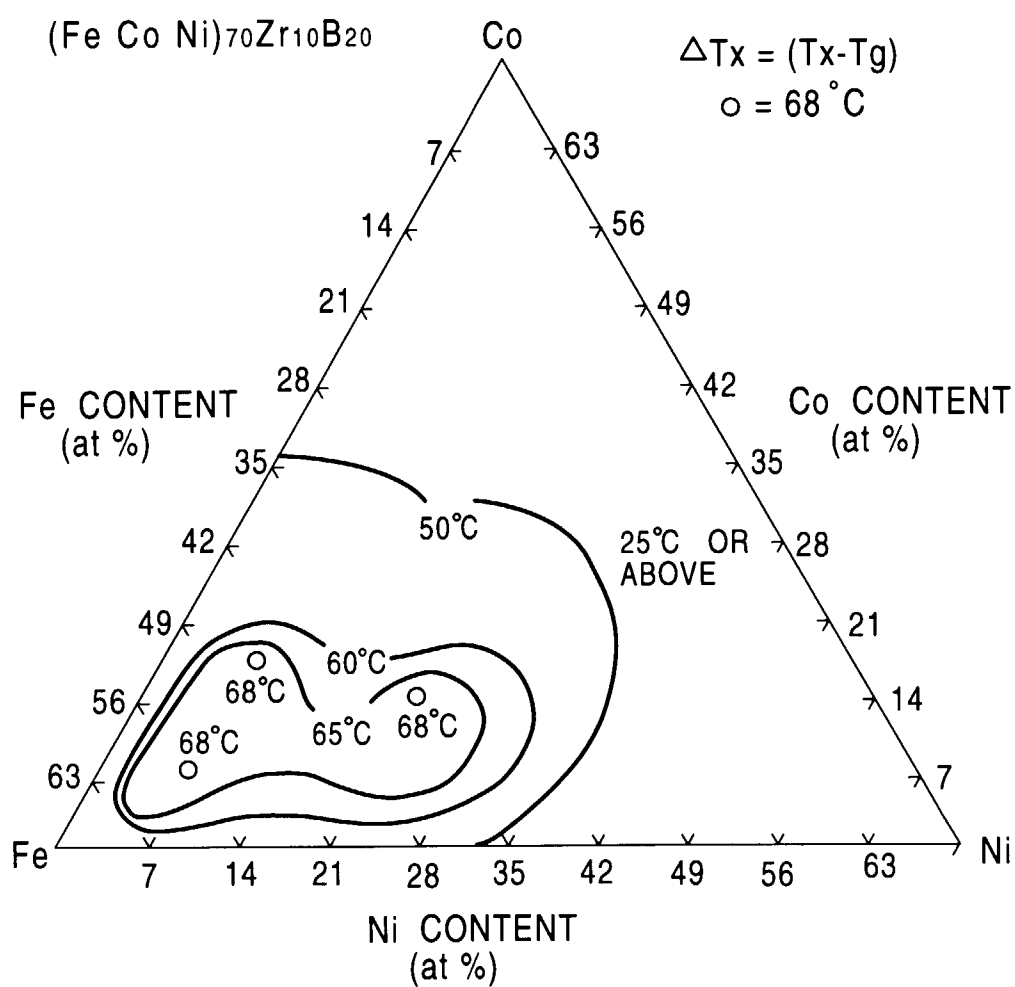


FIG. 8

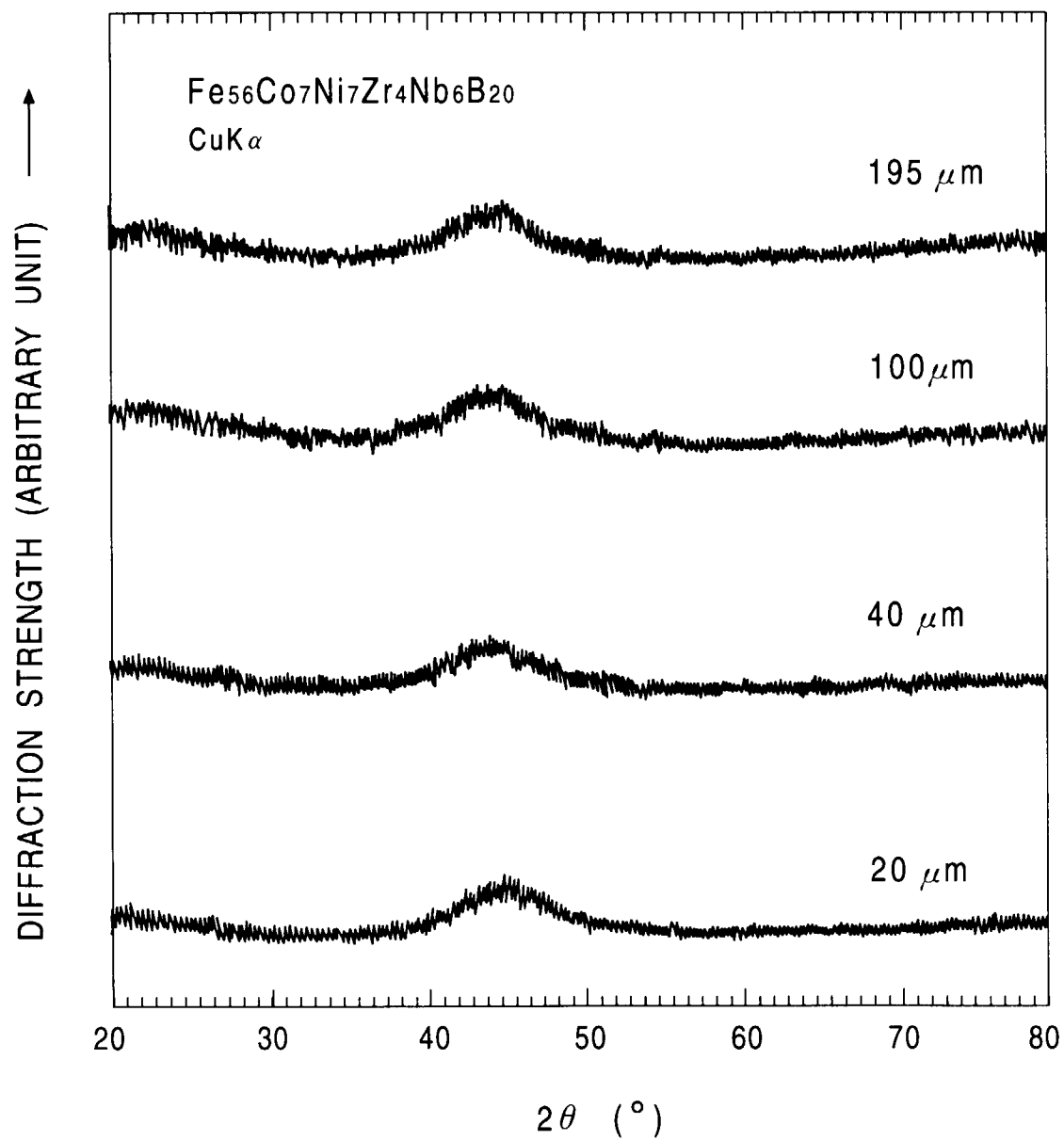


FIG. 9

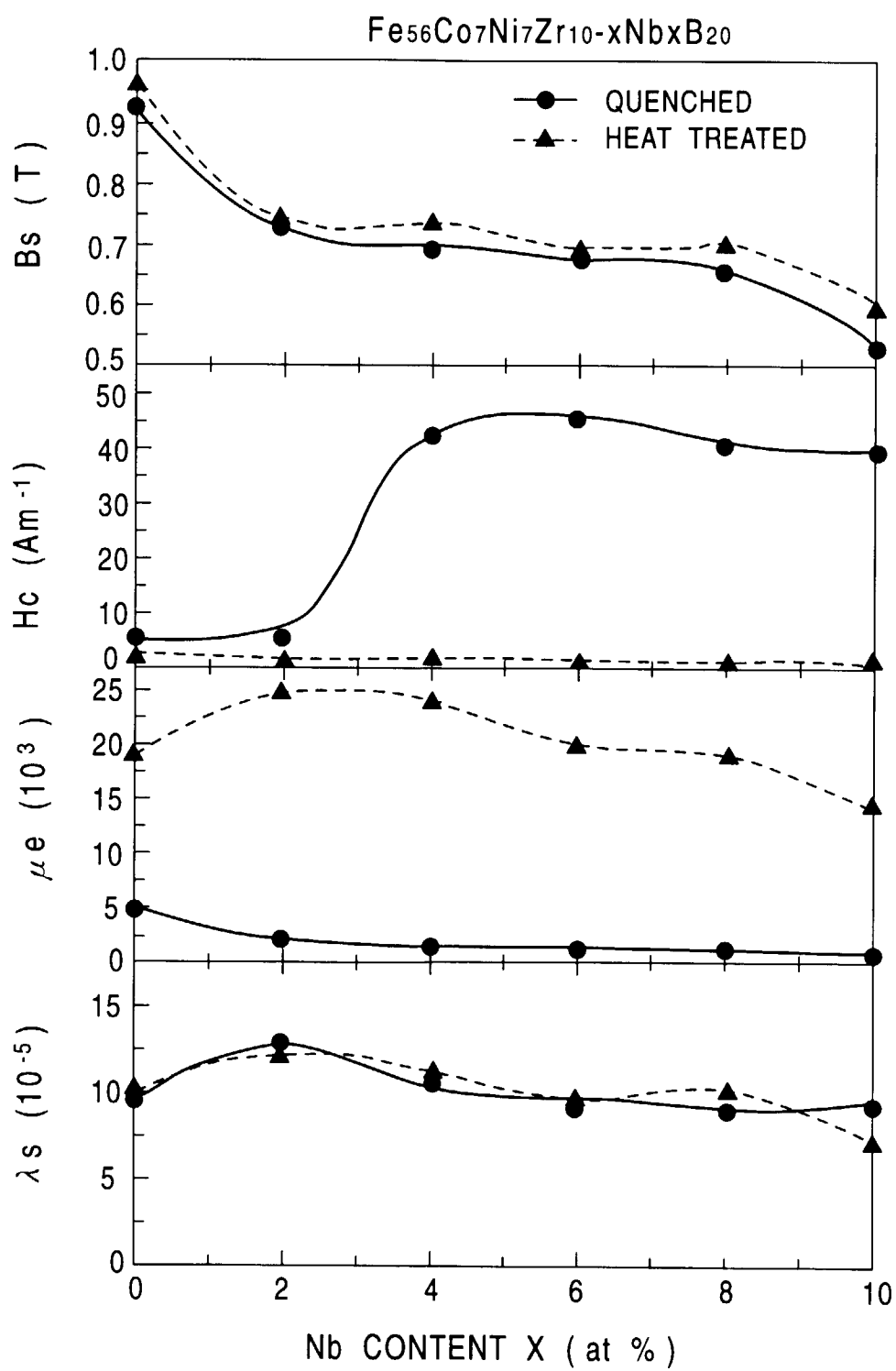


FIG. 10

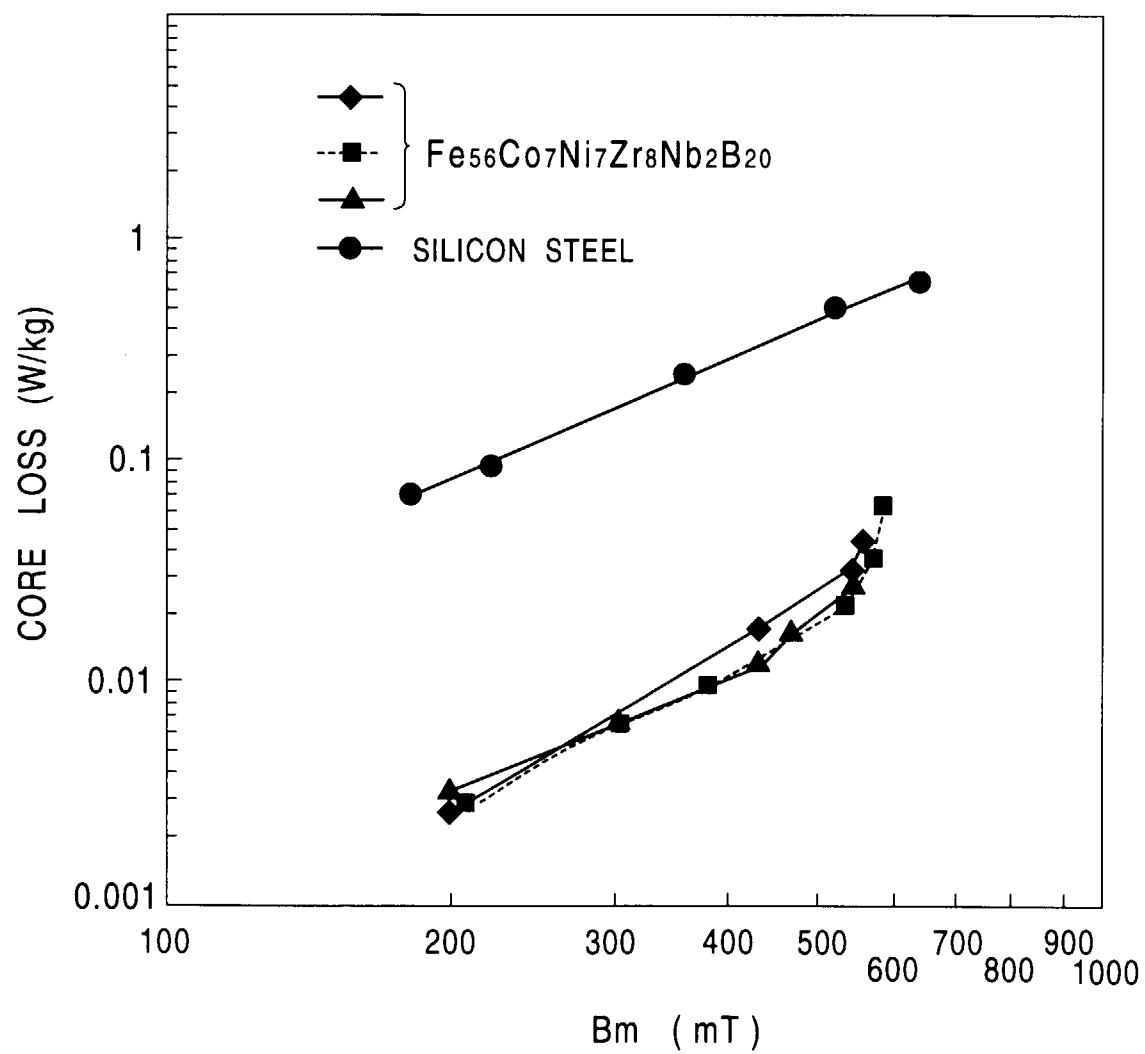


FIG. 11

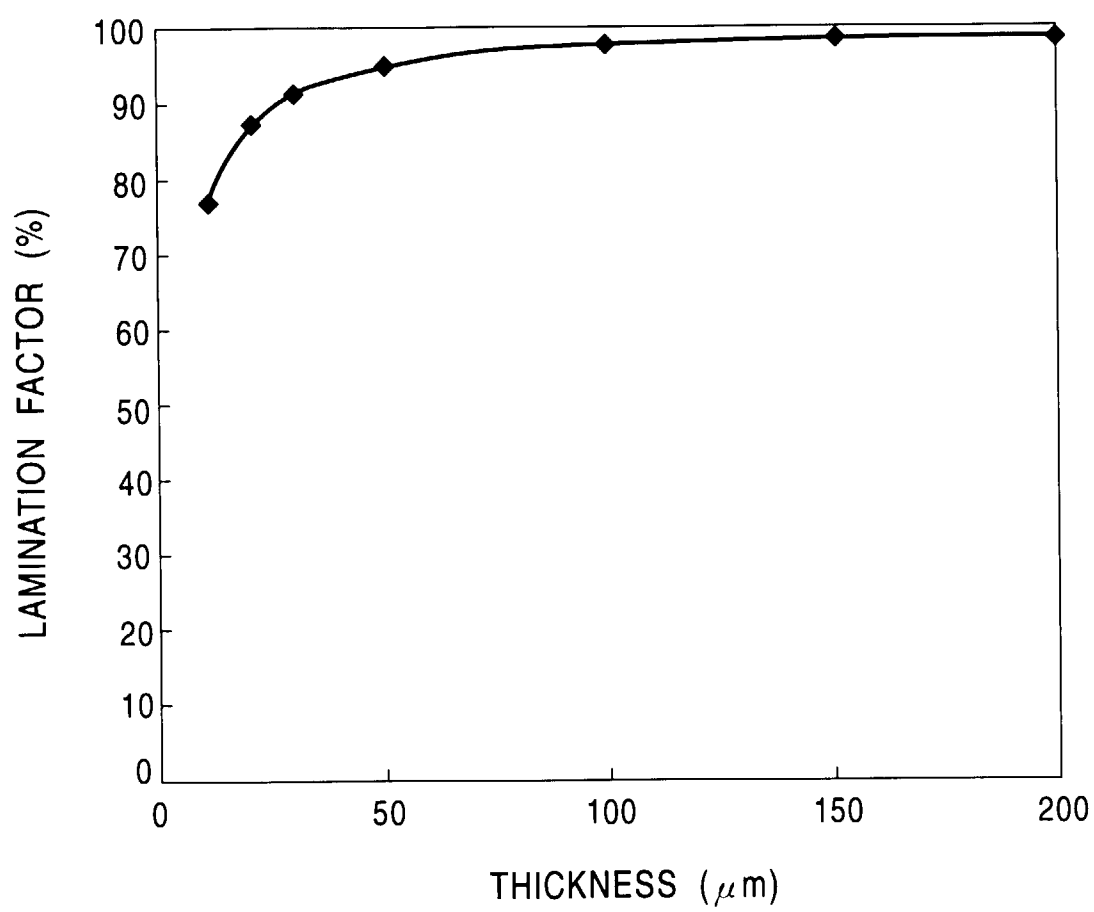


FIG. 12

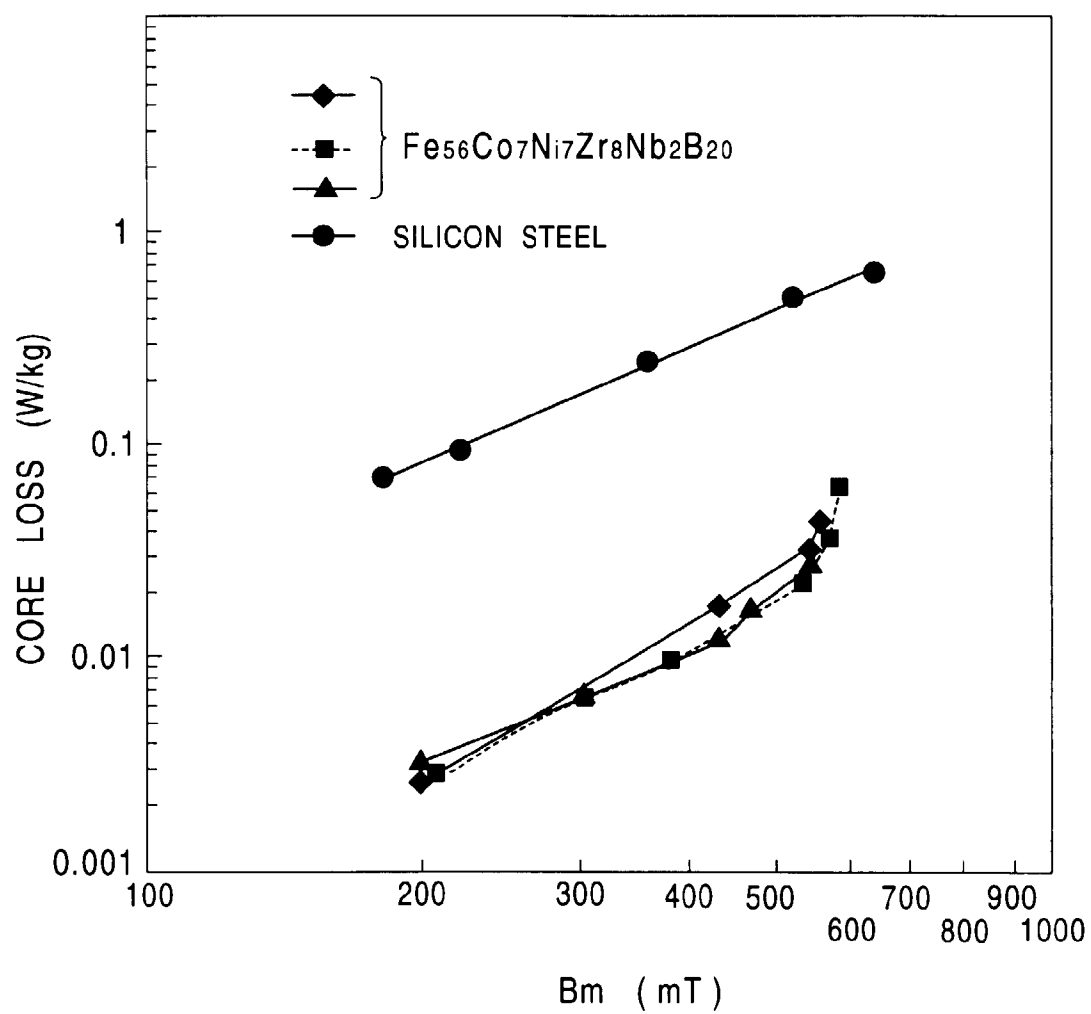
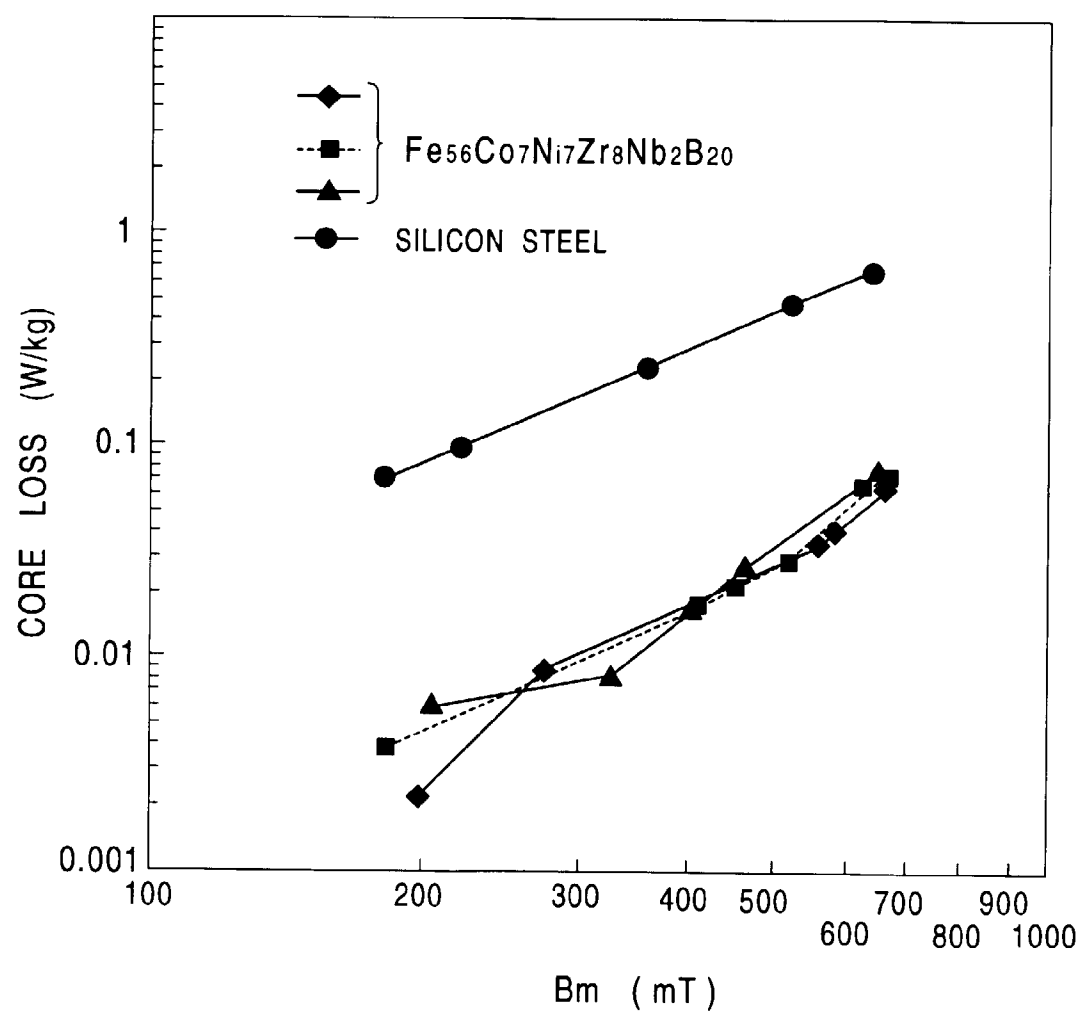


FIG. 13





European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 98 30 6516

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
Y	PATENT ABSTRACTS OF JAPAN vol. 017, no. 442 (C-1097), 16 August 1993 & JP 05 098402 A (NIPPON STEEL CORP), 20 April 1993	1,3,4,6	H01F1/153 H01F41/02 H01F3/04
X	* abstract *	14-17, 20,21	
Y	US 5 509 975 A (KOJIMA AKINORI ET AL) 23 April 1996 * claims 1,5,7,11,13,17; examples 1,3 *	1,3,4,6	
A	EP 0 302 355 A (HITACHI METALS LTD) 8 February 1989 * claims 1,4,5,9,20 *	1,3,4,6, 7,9,11, 12,14	
A	INOUE A ET AL: "BULK AMORPHOUS ALLOYS WITH HIGH MECHANICAL STRENGTH AND GOOD SOFT MAGNETIC PROPERTIES IN FE-TM-B (TM=IV-VIII GROUP TRANSITION METAL) SYSTEM" APPLIED PHYSICS LETTERS, vol. 71, no. 4, 28 July 1997, pages 464-466, XP000698246 * the whole document *	1,3,4,6, 11	TECHNICAL FIELDS SEARCHED (Int.Cl.6) H01F
P,X	DE 198 02 349 A (INOE AKIHISA) 30 July 1998 * page 12, line 50 - page 13, line 2; claims 1-3 *	14,16, 18,20,22	
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 5 November 1998	Examiner Decanniere, L
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

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