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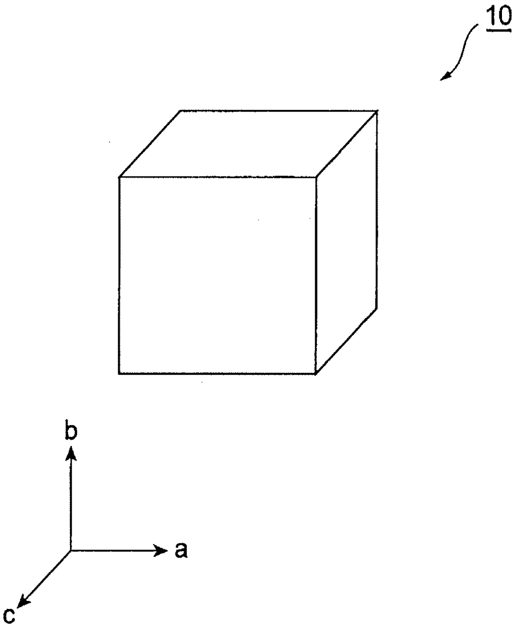
(54) Method of Producing Rare-Earth Magnet

(57) A method of producing a rare-earth magnet containing a rare-earth compound having a first rare-earth element and a second rare-earth element different from the first rare-earth element includes: a mixing step of mixing rare-earth compound powder including the first rare-earth element and subjected to a process based on hydrogenation disproportionation desorption recombination with a diffusion material including the second rare-earth element; a molding step of molding the mixed powder into a compact in a magnetic field; and a heating step of heating the compact to diffuse the second rare-earth element into the rare-earth compound powder.

Fig.1



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Description

BACKGROUND OF THE INVENTION

Field of the Invention

[0001] The present invention relates to a method of producing a rare-earth magnet.

Related Background Art

[0002] Rare-earth bonded magnets have been known as a type of rare-earth magnet containing rare-earth elements. Such rare-earth bonded magnets have excellent magnetic characteristics, and can be relatively easily applied to a complex shape. From this viewpoint, the rare-earth bonded magnets have been used for various apparatuses such as motors. Recently, in various apparatuses, size reduction and efficiency increases have been sought after, and it is necessary to improve magnetic characteristics even further.

[0003] The following method has been proposed as a method of producing a rare-earth bonded magnet. First, magnet powder produced according to the HDDR method (hydrogenation disproportionation desorption recombination) is mixed with diffusion powder including rare-earth elements such as Tb and Dy, and the mixed powder is subjected to a diffusion heat treatment, thereby preparing anisotropic magnet powder in which the rare-earth elements are diffused on the surface of the magnet powder or therein. The anisotropic magnet powder is mixed with resin, coupling agent, antifriction, or the like to produce a rare-earth bonded magnet (e.g., Japanese Patent Publication No. 3452254). In this method of producing the rare-earth bonded magnet, the anisotropic magnet powder in which the rare-earth elements such as Tb and Dy are diffused, is used. Accordingly, it is possible to improve magnetic coercive force and the like.

[0004] However, in the method of producing the rare-earth bonded magnet described in Japanese Patent Publication No. 3452254, the diffusion state of the rare-earth elements in the magnet powder may easily become non-uniform, and the magnetic coercive force or squareness ratio of the obtained rare-earth bonded magnet is not particularly high. In the diffusion heat treatment, it is necessary to heat the magnetic powder at about 700 to 1000°C, and the magnetic powder may be fused to form a mass. For this reason, in this method of producing the rare-earth bonded magnet, even when the magnetic powder having the magnetic anisotropy is used, orientation of the magnetic powder included in the finally obtained rare-earth bonded magnet is disordered and magnetic characteristics such as the squareness ratio deteriorate.

SUMMARY OF THE INVENTION

[0005] An advantage of some aspects of the invention is to provide a method capable of producing a rare-earth magnet having sufficiently excellent magnetic characteristics.

[0006] According to an aspect of the invention, there is provided a method of producing a rare-earth magnet containing a rare-earth compound having a first rare-earth element and a second rare-earth element, including: a mixing step of mixing rare-earth compound powder including the first rare-earth element and subjected to a process based on hydrogenation disproportionation desorption recombination with a diffusion material including the second rare-earth element different from the first rare-earth element; a molding step of molding the mixed powder into a compact in a magnetic field; and a heating step of heating the compact to diffuse the second rare-earth element into the rare-earth compound powder.

[0007] According to the producing method, since the rare-earth compound powder and the diffusion material are heated in close contact with each other, the second rare-earth element included in the diffusion material can be uniformly diffused by outer peripheral portions of the rare-earth compound particles, as compared with a case in which the mixture of the rare-earth compound and the diffusion material mixed in a powder state is subjected to a diffusion process. For this reason, it is possible to produce the rare-earth magnet having the excellent magnetic coercive force (HcJ) and squareness ratio.

[0008] Since the molding is performed in the magnetic field before performing the heating step, it is possible to perform magnetic field orientation in a state where the magnetic powder is not fused. For this reason, the diffusion process is performed in a state where the orientation of the particles of the rare-earth compound powder subjected to the process based on the hydrogenation disproportionation desorption recombination is sufficiently high. Accordingly, it is possible to produce the rare-earth magnet having an excellent residual magnetic flux density (Br) with a high degree of orientation.

[0009] In the producing method of the invention, it is preferable to include an impregnating step of impregnating the compact with resin and curing the resin to obtain the rare-earth bonded magnet after the heating step. Since the rare-earth bonded magnet obtained according to such a producing method is produced using the compact with the high degree of orientation in which the diffusion material is uniformly diffused as described above, the magnetic characteristics thereof are excellent. Since the compact is impregnated with the resin after the producing the compact, it is possible to produce the rare-earth bonded magnet having a high degree of orientation and excellent magnetic characteristics without damaging the orientation of the rare-earth compound powder (HDDR powder) having the magnetic anisotropy, as compared with the case of mixing the resin before producing the compact. According to the method, it is possible to sufficiently reduce variation in size before and after curing the resin, that is, a reduction ratio. For this reason, it is possible to produce the rare-earth bonded magnet with high dimensional precision.

[0010] The producing method of the invention may include, after the heating step, a pulverizing step of pulverizing the compact to prepare pulverized powder and a magnet producing step of molding the mixture of the pulverized powder and resin in the magnetic field and curing the resin to obtain the rare-earth bonded magnet. In the producing method, since the mixture of the pulverized powder and the resin is shaped in the magnetic field, it is possible to easily produce a complex-shaped rare-earth bonded magnet.

[0011] According to the invention, it is possible to provide the method capable of producing the rare-earth magnet having sufficiently excellent magnetic characteristics.

BRIEF DESCRIPTION OF THE DRAWINGS

[0012] Fig. 1 is a perspective view illustrating a rare-earth bonded magnet obtained according to a method of producing a rare-earth magnet according to an embodiment of the invention.

Fig. 2 is a diagram illustrating a magnetic hysteresis loop of the rare-earth bonded magnet obtained according to the method of producing the rare-earth magnet according to the embodiment of the invention.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0013] Hereinafter, exemplary embodiments of the invention will be described with reference to the accompanying drawings.

[0014] A method of producing a rare-earth magnet includes an HDDR step of performing a process based on hydrogenation disproportionation desorption recombination on a raw compound including a first rare-earth element to prepare rare-earth compound powder, a preparing step of preparing a diffusion material including a second rare-earth element, a mixing step of the rare-earth compound powder including the first rare-earth element and subjected to the process based on the hydrogenation disproportionation desorption recombination with the diffusion material including the second rare-earth element to prepare the mixed powder, a molding step of molding the mixed powder into a compact in a magnetic field, a heating step of heating the compact to diffuse the second rare-earth element into outer peripheral portions of the rare-earth compound powder, and an impregnating step of impregnating the compact with resin and curing the resin to obtain a rare-earth bonded magnet. Hereinafter, the processes will be described in detail.

[0015] In the HDDR step, first, the raw compound including the first rare-earth element is prepared. A compound or alloy obtained by a general casting method, for example, a strip casting method, a book molding method, or a centrifugal casting method may be used as the raw compound. A homogenization heat treatment may be further performed. The raw compound may include raw metal, or inevitable impurities derived from the raw compound or the producing method.

[0016] As the first rare-earth element, any rare-earth element may be used, preferably light rare-earth element is used, and more preferably at least one ofNd and Pr is used.

[0017] In the specification, the rare-earth elements are scandium (Sc), yttrium (Y), and lanthanoids belonging to the third group of the long-period form periodic table. Lanthanoids include, for example, lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu). The rare-earth elements can be classified into light rare-earth elements and heavy rare-earth elements. In the specification, the "heavy rare-earth elements" are Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu, and the "light rare-earth elements" are Sc, Y, La, Ce, Pr, Nd, Sm, and Eu.

[0018] A preferable composition of the raw compound may be a R-Fe-B based composition including at least one of Nd and Pr as rare-earth elements, 0.5 to 4.5 mass% of B, and the remainders which are Fe and inevitable impurities. The raw compound may further include another element such as Co, Ni, Mn, Al, Cu, Nb, Zr, Ti, W, Mo, V, Ga, Zn, and Si, as necessary.

[0019] The raw compound having the composition is prepared, and then a process based on the HDDR method is performed. The HDDR method is a process of sequentially performing hydrogenation, disproportionation, desorption, and recombination. The HDDR method will be described in detail hereinafter.

[0020] First, a homogenization heat treatment of keeping the raw compound in the depressurized atmosphere (1 kPa or lower) or the atmosphere of inert gas such as argon and nitrogen at a temperature of 1000 to 1200°C for 5 to 48 hours is performed.

[0021] It is preferable that the homogenized raw compound is pulverized by means such as a stamp mill and a jaw crusher, and then is allowed to pass through a sieve. Accordingly, it is possible to prepare the raw compound in a powder form with a particle size of 10 mm or less.

[0022] In the hydrogen storing step, the raw compound in the powder form is kept in the atmosphere of hydrogen with a hydrogen partial pressure of 100 to 300 kPa at 100 to 200°C for 0.5 to 2 hours. Accordingly, hydrogen is stored in a crystal lattice of the raw material compound.

[0023] Next, the raw compound in which hydrogen is stored, is kept in the atmosphere of hydrogen at a predetermined temperature, thereby performing hydrogenation disproportionation to obtain a decomposition product. It is preferable that the hydrogen partial pressure is 10 to 100 kPa and the temperature is 700 to 850°C at the time of the hydrogenation disproportionation. It is possible to obtain rare-earth compound powder formed from the particles having the magnetic anisotropy by performing the hydrogenation disproportionation under such conditions.

[0024] The decomposition product obtained by the hydrogenation disproportionation includes hydride such as RH_x and iron compounds such as α-Fe and Fe_2B. In this step, the decomposition product has a minute matrix formed therein in the order of 100 nm.

[0025] Subsequently, by reducing the hydrogen partial pressure, hydrogen is discharged from the decomposition product, and anisotropic rare-earth compound powder containing the first rare-earth element is obtained. The rare-earth compound powder has the composition equivalent to that of the raw compound. The particle size of the rare-earth compound powder is preferably 350 μm or less, more preferably 250 μm or less, and even more preferably 212 μm or less. The lower limit of the particle size of the rare-earth compound powder is not particularly limited, but preferably, for example, 1 μm or more.

[0026] The rare-earth compound powder obtained by the HDDR method may be further pulverized using a pulverizing mill such as a jet mill, a ball mill, a vibration mill, and a wet attritor. The rare-earth compound powder subjected to the HDDR method has crystal particles of a small size and is anisotropic. Accordingly, it is possible to easily obtain the rare-earth magnet having a sufficiently high density and excellent magnetic characteristics.

[0027] In the preparing step, the diffusion material in a powder form including the second rare-earth element is prepared. As far as the second rare-earth element is different from the first rare-earth element, the second rare-earth element is not particularly limited. From the viewpoint of obtaining a rare-earth magnet having further high magnetic coercive force, the second rare-earth element is preferably a heavy rare-earth element, and more preferably Dy or Tb. The diffusion material may be a general rare-earth compound such as hydride, oxide, halide, and hydroxide of rare-earth elements, or rare-earth metal. From the view point of further improving the magnetic characteristics of the rare earth magnet, it is preferable to use a heavy rare-earth compound having a heavy rare-earth element as a constituent element.

[0028] The heavy rare-earth compound may include elements other than heavy rare-earth metal elements, and may be alloy of heavy rare-earth metal and metal other than rare-earth metal. From the viewpoint of producing a rare-earth magnet having further excellent magnetic characteristics, the heavy rare-earth compound is preferably hydride and fluoride, and more preferably hydride. When using the heavy rare-earth compound, it is possible to sufficiently reduce the amount of impurities remaining in the rare-earth magnet. Since hydride and fluoride are easily decomposed, it is possible to sufficiently uniformly diffuse the second rare-earth element into the rare-earth compound powder with a minute structure obtained by the HDDR method. By such factors, it is possible to obtain the rare-earth magnet having the further excellent magnetic characteristics. As preferable heavy rare-earth compounds, there are DyH_2, DyF_3, and TbH_2.

[0029] The rare-earth compound or the rare-earth metal can be produced by a general method. Rare-earth compound powder or rare-earth metal powder can be prepared from the rare-earth compound or the rare-earth metal, which is produced by the general method, by a dry pulverizing method using a jet mill, or by mixing with an organic solvent according to a wet pulverizing method using a ball mill or the like.

[0030] An average grain diameter of the diffusion material is preferably 100 nm to 30 μm, more preferably 0.5 to 10 μm, and even more preferably 1 to 5 μm. When the average grain diameter of the diffusion material is more than 30 μm, the second rare-earth element is hardly diffused into the rare-earth compound powder, and the effect of improving the sufficiently large HcJ and squareness ratio may be damaged. Meanwhile, when the average grain diameter of the diffusion material is less than 100 nm, the rare-earth element tends to be easily oxidized. As described above, when the rare-earth oxide is created, the amount of diffusion of the second rare-earth element into the rare-earth compound including the first rare-earth element becomes small, and the improvement of the magnetic coercive force caused by the diffusion tends to decrease. In the specification, an average grain diameter of the diffusion material is a volume average grain diameter (d(50)) measured using a commercially available grain size distribution calculator.

[0031] In the mixing step, the rare-earth compound powder including the first rare-earth element subjected to the HDDR method as described above is mixed with the diffusion material including the second rare-earth element to prepare mixed powder. The mixed powder can be obtained by putting the rare-earth compound powder and the diffusion material into a container, for example, at a predetermined mixing ratio, and then mixing them for 1 to 30 minutes using a SPEX mixer. From the viewpoint of suppressing oxidization of the diffusion material and the rare-earth compound powder, it is preferable to perform the mixing in the atmosphere of inert gas such as argon gas. The mixing method is not particularly limited, and may be a method using, for example, a V mixer, a ball mill, or a Leica device may be used. Antifriction such as zinc stearate being binder for molding at the time of mixing may be added. In this case, the amount of addition thereof may be 0.01 to 0.5 mass%.

[0032] A combination ratio of the rare-earth compound powder and the diffusion material is a ratio in which the content of the diffusion material in the mixed powder is preferably 0.5 to 5 mass%, more preferably 1 to 4 mass%, and even more preferably 1.5 to 3.5 mass%. When the content is less than 0.5 mass%, the amount of diffusion of the second rare-earth element becomes small, and the effect of improving the sufficiently large HcJ and squareness ratio tends to be hardly obtained. Meanwhile, when the content is more than 5 mass%, the second rare-earth element diffused even into the rare-earth compound powder, Br tends to decrease, and costs of materials tend to increase.

[0033] In the molding step, a compact having a desired shape is produced by molding the mixed powder in the magnetic field. The molding in the magnetic field is performed while applying the magnetic field, and the rare-earth compound powder having anisotropy is fixed in a state where it is oriented in a predetermined direction. The molding can be performed, for example, by compression molding using a compression molding machine such as a mechanical press and a hydraulic press. Specifically, a mold cavity is filled therein with the mixed powder, the filled powder is put between an upper punch and a lower punch, and pressure is applied, thereby molding the mixed powder to have a predetermined shape.

[0034] The shape of the compact obtained by the molding is not particularly limited, and is determined according to a shape of a desirable rare-earth bonded magnet such as a pillar shape, a plate shape, and a ring shape. At the time of the molding in the magnetic field, it is preferable to apply the pressure with 580 to 1400 MPa. The orientation magnetic field is preferably 800 to 2000 kA/m. As the molding method, in addition to the dry molding of molding the mixed powder as described above, wet molding using slurry in which the mixed powder is distributed in a solvent such as oil may be applied.

[0035] In the embodiment, the rare-earth compound powder subjected to the HDDR method is used, and the molding of the mixed powder in the magnetic field is performed before the heating step for diffusion without mixing with resin. For this reason, it is possible to make the orientation of the rare-earth compound powder having the magnetic anisotropy sufficiently uniform. Accordingly, it is possible to obtain the rare-earth bonded magnet having particularly an excellent residual magnetic flux density. That is, it is possible to sufficiently exhibit the magnetic characteristics of the rare-earth compound powder having high anisotropy obtained by the HDDR method.

[0036] In the heating step, the compact obtained by the forming in the magnetic field is heated enough to diffuse the second rare-earth element included in the diffusion material into the outer peripheral portions of the rare- earth compound powder. Specifically, the compact is kept under the depressurized condition or under the atmosphere of inert gas such as argon gas, preferably at 700 to 1100 °C, more preferably 700 to 950°C, and even more preferably 800 to 900°C, for 10 minutes to 12 hours. By the heating under such conditions, the second rare-earth element is diffused into the outer peripheral portions of the rare-earth compound powder, and particles having an inner layer in which the first rare-earth element is abundant, and an outer layer in which the second rare-earth element is abundant to coat the inner layer are formed. Accordingly, it is possible to form the rare-earth bonded magnet having sufficiently high magnetic coercive force. In the rare-earth compound powder subjected to the HDDR method, minute cracks exist, but the diffusion material infiltrates the cracks and thus it is possible to fill the cracks with the diffusion material. For this reason, it is possible to improve the oxidization resistance and strength of the finally obtained rare-earth bonded magnet.

[0037] In the heating step, when the heating temperature of the compact is too high or the heating time is too long, the rare-earth compound powder is sintered. In an impregnating step to be performed later, the compact tends to be hardly impregnated therein with resin. Phase decomposition of the anisotropic magnetic powder obtained by performing the HDDR method occurs, and the high magnetic characteristics may be damaged. Meanwhile, when the heating temperature of the compact is too low or the heating time is too short, the diffusion of the second rare-earth element tends not to sufficiently proceed. Accordingly, it is preferable to set the heating temperature and the heating time according to the kinds of the first and second rare-earth elements or the particle size of the rare-earth compound powder.

[0038] In the producing method of the embodiment, since the heating step is performed in the state where rare-earth compound powder is made into the compact, adhesion of the rare-earth compound powder and the diffusion material is satisfactory and it is possible to more uniformly diffuse the second rare-earth element into the outer peripheral portions of the rare-earth compound powder. Accordingly, it is possible to obtain the rare-earth bonded magnet with the sufficiently high squareness ratio and magnetic coercive force.

[0039] In the impregnating step, the compact is impregnated with resin and is heated to harden the resin, and a rare-earth bonded magnet containing the rare-earth compound particles and the resin filling the spaces among the rare-earth compound particles is obtained. Specifically, first, the compact subjected to the heating step is immersed in a resin-contained solution prepared in advance, and is defoamed by depressurizing an air-tight container, thereby putting the resin-contained solution into the voids of the compact. Then, the compact is taken out of the resin-contained solution, and the residual resin-contained solution attached to the surface of the compact is removed. A centrifugal separator or the like may be used to remove the residual resin-contained solution. Before the compact is immersed in the resin-contained solution, the compact is put into the air-tight container and is immersed in a solvent such as toluene while keeping the container in the depressurized atmosphere, thereby promoting defoaming. Accordingly, it is possible to increase the amount of impregnation, and it is possible to reduce the voids in the compact.

[0040] The resin contained in the resin-contained solution may be thermosetting resin such as epoxy resin and phenol resin, and thermoplastic resin such as styrene-based, olefin-based, urethane-based, polyester-based, polyamide-based elastomer such as nylon, ionomer, ethylene propylene copolymer (EPM), and ethylene-ethyl acrylate copolymer. The thermosetting resin is preferable among them, and the epoxy resin or phenol resin is more preferable.

[0041] The resin-contained solution can be prepared by dissolving the resin into a solvent. As the solvent, a general organic solvent such as toluene, acetone, and ethyl alcohol may be used. It is preferable to select the solvent according to the kinds of used resin to sufficiently dissolve the resin. The resin content of the resin-contained solution is not particularly limited, and it is preferable that the resin content is large to obtain a rare-earth bonded magnet with high density and few voids.

[0042] The compact in which the resin-contained solution is infiltrated into the voids is kept for example, in a thermostatic chamber in the depressurized atmosphere (1 kPa or less) or the atmosphere of inert gas such as argon gas and nitrogen gas at 120 to 230°C for 1 to 5 hours. Thus, the solvent included in the resin-contained solution is evaporated and the resin is cured. Then, a surface treating is performed as necessary, and it is possible to obtain the anisotropic rare-earth bonded magnet.

[0043] The resin content of the rare-earth bonded magnet is preferably 0.5 to 10 mass%, and more preferably 1 to 5 mass% from the viewpoint of achieving both excellent magnetic characteristics and an excellent shape-keeping property. The amount of impregnated resin can be controlled by changing the concentration of the resin in the resin-contained solution or the molding pressure at the time of producing the compact.

[0044] Fig. 1 is a perspective view illustrating the rare-earth bonded magnet obtained according to the producing method of the embodiment. The rare-earth bonded magnet 10 obtained according to the producing method of the embodiment contains the particles having the rare-earth compound as a main component and the resin filling the spaces among the particles. The rare-earth compound includes the first rare-earth element and the second rare-earth element as constituent elements. In the rare-earth bonded magnet 10, the second rare-earth element included in the diffusion material is uniformly diffused into the outer peripheral portions of the rare-earth compound particles as compared with the known magnet, and thus the rare-earth bonded magnet 10 has the sufficiently high HcJ and squareness ratio. The anisotropic rare-earth compound powder is shaped in the magnetic field without the heating step and mixing with resin. Accordingly, it is possible to make the orientation of the particles of the rare-earth compound more uniform than the known technique, the degree of orientation becomes high, and it is possible to produce the rare-earth bonded magnet with excellent magnetic characteristics.

[0045] A modified example of the embodiment will be described. A method of producing a rare-earth magnet according to the modified example includes a pulverizing step of pulverizing the compact obtained in the heating step and formed of the rare-earth compound powder in which the second rare-earth element is diffused into the outer peripheral portions to prepare pulverized powder, and a magnet producing step of molding a mixture of the pulverized powder and resin in the magnetic field and curing the resin to obtain the rare-earth bonded magnet.

[0046] The producing method according to the modified example is the same as the embodiment from the HDDR step of producing the compact to the heating step of diffusing the second rare-earth element included in the diffusion material into the rare-earth compound powder.

[0047] In the pulverizing step, the compact is pulverized by a stamp mill, and is allowed to pass through a sieve as necessary, thereby preparing pulverized powder with an average particle size of 100 to 300 μm. The pulverizing method is not particularly limited, and a jaw crusher or various kinds of the known pulverizing means may be used.

[0048] In the magnet producing step, first, a bonded magnet compound that is a mixture of the pulverized powder and the resin is prepared. Specifically, the pulverized powder is mixed with the same resin-contained solution as that of the embodiment and is heated to volatilize at least a part of the solvent of the resin-contained solution, thereby obtaining the bonded magnet compound. The resin content of the bonded magnet compound is preferably 0.5 to 10 mass%, more preferably 1 to 5 mass%, and even more preferably 1 to 3 mass%.

[0049] Next, the bonded magnet compound is shaped in the magnetic field to produce a compact. The molding in the magnetic field may be performed in the same manner as the embodiment. Thereafter, the produced compact is kept for example, in a thermostatic chamber in the depressurized atmosphere (1 kPa or less) or the atmosphere of inert gas such as argon gas and nitrogen gas at 120 to 230°C for 1 to 5 hours. Thus, the solvent is evaporated and the resin is cured. Then, a surface treating is performed as necessary, and it is possible to obtain the rare-earth bonded magnet 10 shown in Fig. 1.

[0050] In the producing method according to the modified example, since the heating step is performed on the compact in the state after the molding in the magnetic field in the same manner as the embodiment, the second rare-earth element included in the diffusion material is more uniformly diffused than the known method. Accordingly, it is possible to obtain the rare-earth bonded magnet having the sufficiently high HcJ and squareness ratio. Since the molding can be performed after mixing the pulverized powder including the rare-earth compound powder and the diffusion material with the resin-contained solution, it is possible to easily produce a rare-earth bonded magnet with a complex shape.

[0051] Preferable embodiments have been described above, but the invention is not limited to the embodiments. In the embodiment, the rare-earth bonded magnet is exemplified as the rare-earth magnet, but the rare-earth magnet obtained according to the producing method of the invention may have an aspect in which it is not impregnated with resin. Such a rare-earth magnet may be, for example, anything which can be obtained by the heating step of the embodiment.

Examples

[0052] The contents of the invention will be described hereinafter with reference to Examples and Comparative Examples, but the invention is not limited to the following examples.

(Example 1)

<Production of Rare-Earth Bonded Magnet>

[0053] A raw compound containing $Nd_2Fe_{14}B$ as a main component and having the following composition was prepared according to a strip casting method.
[0054]

Nd: 28.0 mass%
B: 1.1 mass%
Ga: 0.35 mass%
Nb: 0.30 mass%
Cu: 0.03 mass%
Co: 3.8 mass%
Fe and Inevitable Impurities: Remainder

[0055] The raw compound included a small amount of inevitable impurities (0.5 mass% or less in the whole raw compound). The raw compound was kept in the depressurized atmosphere (1 kPa or lower) in the temperature range of 1000 to 1200°C for 24 hours (homogenization heat treatment process). The product ($Nd_2Fe_{14}B$) obtained by the homogenization heat treatment was pulverized using a stamp mill and was allowed to pass through a sieve, and raw powder (particle size: 1 to 2 mm) was obtained.

[0056] The raw powder was put into a container made of molybdenum, and a tubular heating furnace with an infrared heating manner was loaded with the container. Then, a process based on the hydrogenation disproportionation desorption recombination (HDDR method) was performed.

[0057] First, a hydrogen storing step of keeping the raw powder in the atmosphere of hydrogen gas under a hydrogen partial pressure of 100 to 300 kPa at a temperature of 100° for 2 hours was performed. Subsequently, the hydrogen partial pressure in the furnace was lowered while the temperature in the furnace was raised, and a hydrogenation disproportionation step of keeping the raw powder in which hydrogen gas is stored, under the conditions of a hydrogen partial pressure of 40 kPa and a temperature of 850°C for 1.5 hours was performed.

[0058] Thereafter, a desorption recombination step was performed by lowering the hydrogen pressure while the inside of the furnace was kept at 850°C. Thus, anisotropic magnetic powder subjected to the HDDR method was obtained. The obtained magnetic powder was pulverized using a stamp mill in the atmosphere of nitrogen gas, and was allowed to pass through a sieve, thereby obtaining $Nd_2Fe_{14}B$ powder with a particle size of 300 μm or less.

[0059] Subsequently, a diffusion material was prepared as follows, separately from the $Nd_2Fe_{14}B$ powder. First, hydrogen was stored in Dy powder in the atmosphere of hydrogen at 350°C for 1 hour, and then the Dy powder was processed in the atmosphere of Ar at 600°C for 1 hour, thereby obtaining Dy hydride. The obtained Dy hydride was confirmed as DyH_2 by X-ray diffraction measurement. The obtained DyH_2 powder was put into an ethanol solution and was subjected to a ball mill pulverizing process, and it was made into fine DyH_2 powder with an average particle size (d (50)) of 3 μm.

[0060] The $Nd_2Fe_{14}B$ powder obtained according to the above-described method was mixed with the fine DyH_2 powder as the diffusion material using a V mixer, thereby preparing mixed powder. A mixing ratio of the $Nd_2Fe_{14}B$ powder and the diffusion material was a ratio in which the diffusion material was 3 mass% when the whole obtained powder was a reference. Zinc stearate was added at a concentration of 0.1 mass % and was mixed into the whole amount of the mixed powder. The mixed powder was shaped in the magnetic field under the conditions of a molding pressure of 980 MPa and an orientation magnetic field of 1.2 T, and a rectangular parallelepiped compact 10 shown in Fig. 1 was obtained. A magnetic field applying direction was a direction indicated by the arrow a shown in Fig. 1. The dimension and density of the compact 10 are as shown in Table 1.

[0061] The compact was subjected to a diffusion process of diffusing the Dy included in the diffusion material into the outer peripheral portions of the $Nd_2Fe_{14}B$ powder by a heat treatment of heating the compact in the atmosphere of argon gas at 900°C for 30 minutes. A relative density of the compact after the diffusion process was about 80%.

[0062] Next, the compact was put into a vacuum bell jar with the container in which toluene was put, the compact was deposited into the toluene, a defoaming process of keeping it in a state where the pressure in the container is 10 kPa or less for 30 minutes was performed, and the pressure was returned to a normal pressure.

[0063] Epoxy resin was dissolved into the toluene to prepare an epoxy resin solution (the content of epoxy resin: 50 mass%), separately from the compact. The epoxy resin solution and the compact subjected to the defoaming process by performing the diffusion process are sequentially put into the vacuum bell jar. The inside of the vacuum bell jar was depressurized to 10 kPa or lower and was kept for 60 minutes, and thus the epoxy resin solution was infiltrated into the compact.

[0064] The compact was taken out of the epoxy resin solution, and the epoxy resin solution attached to the surface of the compact was removed by a centrifugal separator. Thereafter, the compact impregnated with the epoxy resin solution was kept in the thermostatic chamber at a temperature of 150°C (atmosphere: nitrogen gas) for 5 hours, and the epoxy resin in the compact was cured, thereby obtaining the rare-earth bonded magnet 10. The dimension and mass of the obtained rare-earth bonded magnet 10 were measured to calculate the density of the rare-earth bonded magnet. The dimensions and density of the rare-earth bonded magnet are shown in Table 1 and Table 2.

[0065]

[Table 1]

	Length of One Side (mm)			Density
Direction (Note 1)	a	b	c	(g/cm^3)
Compact α before Diffusion Process	11.102	11.367	10.114	5.900
Rare-Earth Bonded Magnet β	11.065	11.315	10.019	5.987
Variation in Size (β-α)/β	-0.33%	-0.46%	-0.94%	
Note 1: Directions are the directions a, b, and c shown in Fig. 1 Note 2: Magnetic field applying direction at the time of shaping in the magnetic field is the direction a shown in Fig. 1				

<Measurement of Magnetic Characteristics>

[0066] The magnetic characteristics of the rare-earth bonded magnet produced as described above were measured by a BH tracer. A residual magnetic flux density (Br), a coercive magnetic force (HcJ), and a squareness ratio (Hk/iHc) were calculated from the obtained result. Hk and bHc were calculated from a magnetic hysteresis loop, and the squareness ratio was calculated using HcJ and Hk on the basis of the following formula (1). The measurement result is shown in Table 2.

[0067] The squareness ratio is an indicator of magnetic performance, and denotes an angular degree in the second quadrant of the magnetic hysteresis loop measured using the BH tracer. The Hk of the formula (1) denotes an external magnetic intensity when a ratio of magnetization to the residual magnetic flux density becomes 90% with respect to the second quadrant of the magnetic hysteresis loop.

[0068]

$$\text{Squareness Ratio (\%)} = Hk/HcJ \times 100 \qquad (1)$$

(Example 2)

[0069] The rare-earth bonded magnet was prepared and measured in the same manner as Example 1, except for changing the mixing ratio of the diffusion material (fine DyH_2 powder) from 3 mass% to 1 mass% when the whole mixed powder was a reference. The measurement result is shown in Table 2.

(Example 3)

[0070] The diffusion process of the compact was performed in the same manner as Example 1. After the diffusion process, the compact was pulverized using a stamp mill in the atmosphere of nitrogen gas, to prepare the pulverized powder with a particle size of 250 μm or less. The pulverized powder was mixed with the epoxy resin solution prepared in the same manner as Example 1 at a ratio in which the content of the epoxy resin of the compact was 3 mass%, toluene was evaporated, and a bonded magnet compound formed of magnetic powder and resin was prepared. The bonded magnet compound was shaped in the magnetic field under the conditions of a molding pressure of 980 MPa and an orientation magnetic field of 1.2 T, and a rectangular parallelepiped compact was obtained.

[0071] The compact was kept in the thermostatic chamber at a temperature of 150°C for 5 hours, and the epoxy resin in the compact was cured, thereby obtaining a rare-earth bonded magnet. The obtained rare-earth bonded magnet was measured in the same manner as Example 1. The measurement result is shown in Table 2.

(Example 4)

[0072] A rare-earth bonded magnet was prepared and measured in the same manner as Example 1, except for using fine DyH_2 powder with an average particle size of 1 μm instead of the fine DyH_2 powder with an average particle size of 3 μm. The measurement result is shown in Table 2. The average particle size of the fine DyH_2 powder was controlled by changing the conditions of the ball mill pulverization.

(Example 5)

[0073] A rare-earth bonded magnet was prepared and measured in the same manner as Example 1, except for using fine powder of Dy-Fe compound (Dy:Fe = 80:20 (mole ratio)) with an average particle size of 3 μm instead of the fine DyH_2 powder as the diffusion material. The measurement result is shown in Table 2. The fine powder of the Dy-Fe compound was prepared as follows. First, DyFe alloy and Fe (electrolytic iron) were weighed and combined to be a desirable composition and were dissolved by high-frequency dissolution, and a Dy-Fe compound was obtained according to a strip casting method. The obtained Dy-Fe compound was pulverized using a jaw crusher, and then was pulverized using a ball mill in an ethanol solution at 100 rpm for 120 hours, thereby obtaining fine powder of the Dy-Fe compound with an average particle size of 3 μm.

(Example 6)

[0074] A rare-earth bonded magnet was prepared and measured in the same manner as Example 1, except for using fine powder of a Dy-Fe-Co compound (Dy:Fe:Co = 80:10:10 (mole ratio)) with an average particle size of 3 μm instead of the fine DyH_2 powder as the diffusion material. The measurement result is shown in Table 2. The fine powder of the Dy-Fe-Co compound was prepared as follows. First, DyFe alloy, Fe (electrolytic iron), and Co were weighed and combined to be a desirable composition and were melted by arc melting and solidified, thereby obtaining a Dy-Fe-Co compound. The Dy-Fe-Co compound was coarsely pulverized using a stamp mill in the atmosphere of nitrogen, and then was pulverized using a ball mill in an ethanol solution at 100 rpm for 60 hours, thereby obtaining fine powder of the Dy-Fe-Co compound with an average particle size of 3 μm.

(Example 7)

[0075] A rare-earth bonded magnet was prepared and measured in the same manner as Example 1, except for using fine DyF_3 powder with an average particle size of 3 μm instead of the fine DyH_2 powder as the diffusion material. The measurement result is shown in Table 2. The fine DyF_3 powder was prepared by pulverizing DyF_3 powder made by Nihon Yttrium Co., Ltd. using a ball mill in an ethanol solution at 100 rpm for 12 hours.

(Example 8)

[0076] The rare-earth bonded magnet was prepared and measured in the same manner as Example 7, except for changing the mixing ratio of the diffusion material (fine DyF_3 powder) from 3 mass% to 1 mass% when the whole mixed powder was a reference. The measurement result is shown in Table 2.

(Comparative Example 1)

[0077] Mixed powder of $Nd_2Fe_{14}B$ powder and a diffusion material (fine DyH_2 powder) was obtained in the same manner as Example 1. The mixed powder was subjected to a diffusion process of diffusing the Dy included in the diffusion material into the $Nd_2Fe_{14}B$ powder by a heat treatment of heating the mixed powder in the atmosphere of argon gas at 900°C for 30 minutes.

[0078] The mixed powder subjected to the diffusion process was mixed with the epoxy resin solution in the same manner as Example 1, toluene was evaporated, the bonded magnet compound formed of the magnetic powder and the resin is adjusted, and the compound was shaped in the magnetic field under the conditions of a molding pressure of 980 MPa and an orientation magnetic field of 1.2 T, thereby obtaining a compact.

[0079] The compact was kept in the thermostatic chamber at a temperature of 150°C for 5 hours, and the epoxy resin in the compact was cured, thereby obtaining a rare-earth bonded magnet. The obtained rare-earth bonded magnet was measured in the same manner as Example 1. The measurement result is shown in Table 2.

(Comparative Example 2)

[0080] A rare-earth bonded magnet was produced and measured in the same manner as Comparative Example 1, except for changing the content of the fine DyH_2 powder from 3 mass% to 1 mass% in the mixed powder. The measurement result is shown in Table 2.

(Comparative Example 3)

[0081] A rare-earth bonded magnet was produced and measured in the same manner as Example 1 except for using only the $Nd_2Fe_{14}B$ powder instead of the mixed powder of the $Nd_2Fe_{14}B$ powder and diffusion material (fine DyH_2 powder). The measurement result is shown in Table 2.

(Comparative Example 4)

[0082] $Nd_2Fe_{14}B$ powder with a particle size of 300 μm or less was obtained in the same manner as Example 1. The epoxy resin solution prepared in the same manner as Example 1 was mixed with the $Nd_2Fe_{14}B$ powder at a ratio in which the content of the epoxy resin of the compact was 3 mass%, toluene was evaporated, and a bonded magnet compound formed of magnetic powder ($Nd_2Fe_{14}B$ powder) and resin was prepared. The bonded magnet compound was shaped in the magnetic field under the conditions of a molding pressure of 980 MPa and an orientation magnetic field of 1.2 T, and a rectangular parallelepiped compact was obtained.

[0083] The compact was kept in the thermostatic chamber at a temperature of 150°C for 5 hours, and the epoxy resin in the compact was cured, thereby obtaining a rare-earth bonded magnet. The obtained rare-earth bonded magnet was measured in the same manner as Example 1. The measurement result is shown in Table 2.

(Comparative Example 5)

[0084] An ingot with a predetermined composition was melted in a high-frequency melting furnace, was pulverized using a stamp mill in the atmosphere of nitrogen gas, and allowed to pass through a sieve, thereby obtaining $Nd_2Fe_{14}B$ powder with a grain diameter of 300 μm or less. A rare-earth bonded magnet was produced and measured in the same manner as Example 1 except for using the $Nd_2Fe_{14}B$ powder which was not subjected the HDDR method instead of the $Nd_2Fe_{14}B$ powder subjected to the HDDR method. The measurement result is shown in Table 2.

[0085]

[Table 2]

	Diffusion Material			Hcj (kOe)	Br (kG)	Hk (kOe)	Squareness Ratio Hk/Hcj (%)	Density (g/cm^3)
	Kind	Average Particle Size (μm)	Mixing Ratio (mass%)					
Ex.1	DyH_2	3	3	20.10	8.74	10.06	50.0	5.99
Ex.2	DyH_2	3	1	18.00	8.70	9.27	51.5	5.96
Ex.3	DyH_2	3	3	19.98	8.56	7.75	38.8	5.93
Ex.4	DyH_2	1	3	20.50	8.69	10.97	53.5	5.95

(continued)

	Diffusion Material			Hcj (kOe)	Br (kG)	Hk (kOe)	Squareness Ratio Hk/Hcj (%)	Density (g/cm^3)
	Kind	Average Particle Size (μm)	Mixing Ratio (mass%)					
Ex.5	80Dy-20Fe	3	3	18.90	8.80	8.79	46.5	5.97
Ex.6	80Dy-10Fe-10Co	3	3	19.30	8.78	9.55	49.5	5.98
Ex.7	DyF_3	3	3	19.00	8.78	9.42	49.6	5.97
Ex.8	DyF_3	3	1	17.05	8.91	8.15	47.8	5.94
Comp.1	DyH_2	3	3	18.60	8.52	5.21	28.0	5.96
Comp.2	DyH_2	3	1	16.90	8.45	4.95	29.3	5.92
Comp.3	-	-	0	15.40	8.94	4.17	27.1	5.82
Comp.4	-	-	0	16.00	8.67	5.23	32.7	5.70
Comp.5	DyH_2	1	3	0.05	2.10	0.01	20.0	5.95

[0086] As shown in Table 2, in Examples 1 to 8 subjected to the diffusion process of the diffusion material in the compact state, the magnetic characteristics were higher than those of Comparative Examples 1 and 2 subjected to the diffusion process of the diffusion material in the powder state. Particularly, the magnetic coercive force and the squareness ratio were drastically improved. In Comparative Examples 3 and 4 which were not subjected to the diffusion process, the magnetic characteristics were lower than those of Examples 1 to 8. In Comparative Example 5 using the rare-earth compound powder which was not subjected to the HDDR method, the magnetic characteristics were significantly lower than those of Examples 1 to 8. The reason seems to be that the magnetic coercive force is not improved even when the diffusion material is diffused into the rare-earth compound powder, since the rare-earth compound powder ($Nd_2Fe_{14}B$) which is not subjected to the HDDR method is not anisotropic powder formed with a microstructure.

[0087] Fig. 2 is a diagram illustrating a magnetic hysteresis loop measured using a BH tracer. In Fig. 2, a curve 1 is a magnetic hysteresis loop of the rare-earth bonded magnet of Example 1, and a curve 2 is a magnetic hysteresis loop of the rare-earth bonded magnet of Comparative Example 1. It was confirmed that the squareness property of the rare-earth bonded magnet of Example 1 is more excellent than that of Comparative Example 1.

Claims

1. A method of producing a rare-earth magnet containing a rare-earth compound having a first rare-earth element and a second rare-earth element different from the first rare-earth element, comprising:

a mixing step of mixing rare-earth compound powder including the first rare-earth element and subjected to a process based on hydrogenation disproportionation desorption recombination with a diffusion material including the second rare-earth element;
a molding step of molding the mixed powder into a compact in a magnetic field; and
a heating step of heating the compact to diffuse the second rare-earth element into the rare-earth compound powder.

2. The method of producing the rare-earth magnet according to Claim 1, further comprising an impregnating step of impregnating the compact with resin and curing the resin to obtain the rare-earth bonded magnet after the heating step.

3. The method of producing the rare-earth magnet according to Claim 1, further comprising:

a pulverizing step of pulverizing the compact to prepare pulverized powder after the heating step; and a magnet producing step of molding the mixture of the pulverized powder and resin in the magnetic field and curing the resin to obtain the rare-earth bonded magnet.

Amended claims in accordance with Rule 137(2) EPC.

1. A method of producing a rare-earth magnet containing a rare-earth compound including a first rare-earth element and a second rare earth element different from the first rare-earth element comprising:

a mixing step of mixing rare-earth compound powder including the first rare-earth element and subjected to a process based on hydrogenation disproportionation desorption recombination with a diffusion material including the second rare-earth element;
a molding step of molding the mixed powder into a compact in a magnetic field; and
a heating step of heating the compact to diffuse the second rare-earth element into the rare-earth compound powder,
wherein
the first rare-earth element is a light rare earth element, and
the diffusion material is selected from the group consisting of a hydride, an oxide, a halide and a hydroxide of a rare-earth element, a rare-earth metal, and an alloy of a heavy rare-earth metal and a metal other than a rare earth metal.

2. The method of producing a rare-earth magnet according to claim 1, wherein the rare-earth compound including a first rare-earth element is prepared from a raw compound which is a R-Fe-B based composition including at least one of Nd and Pr as rare-earth elements, 0.5 to 4.5 mass% of B, and the remainders which are Fe and inevitable impurities.

3. The method of producing a rare-earth magnet according to claim 1 or 2, wherein the second rare-earth element is a heavy rare-earth element.

4. The method of producing the rare-earth magnet according to claim 1, further comprising an impregnating step of impregnating the compact with resin and curing the resin to obtain the rare-earth bonded magnet after the heating step.

5. The method of producing the rare-earth magnet according to claim 1, further comprising:

a pulverizing step of pulverizing the compact to prepare pulverized powder after the heating step; and
a magnet producing step of molding the mixture of the pulverized powder and resin in the magnetic field and curing the resin to obtain the rare-earth bonded magnet.

Fig.1

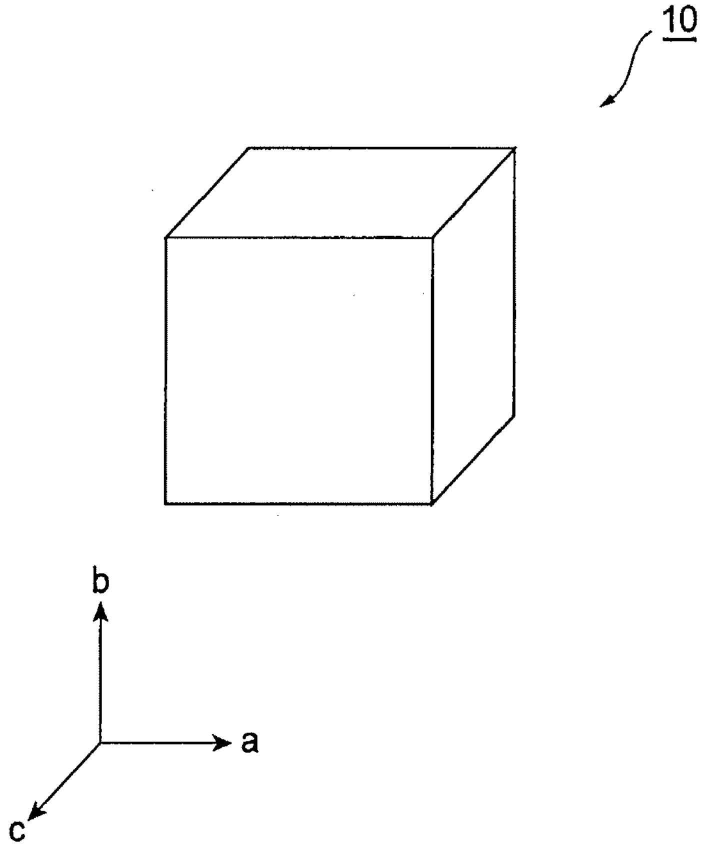
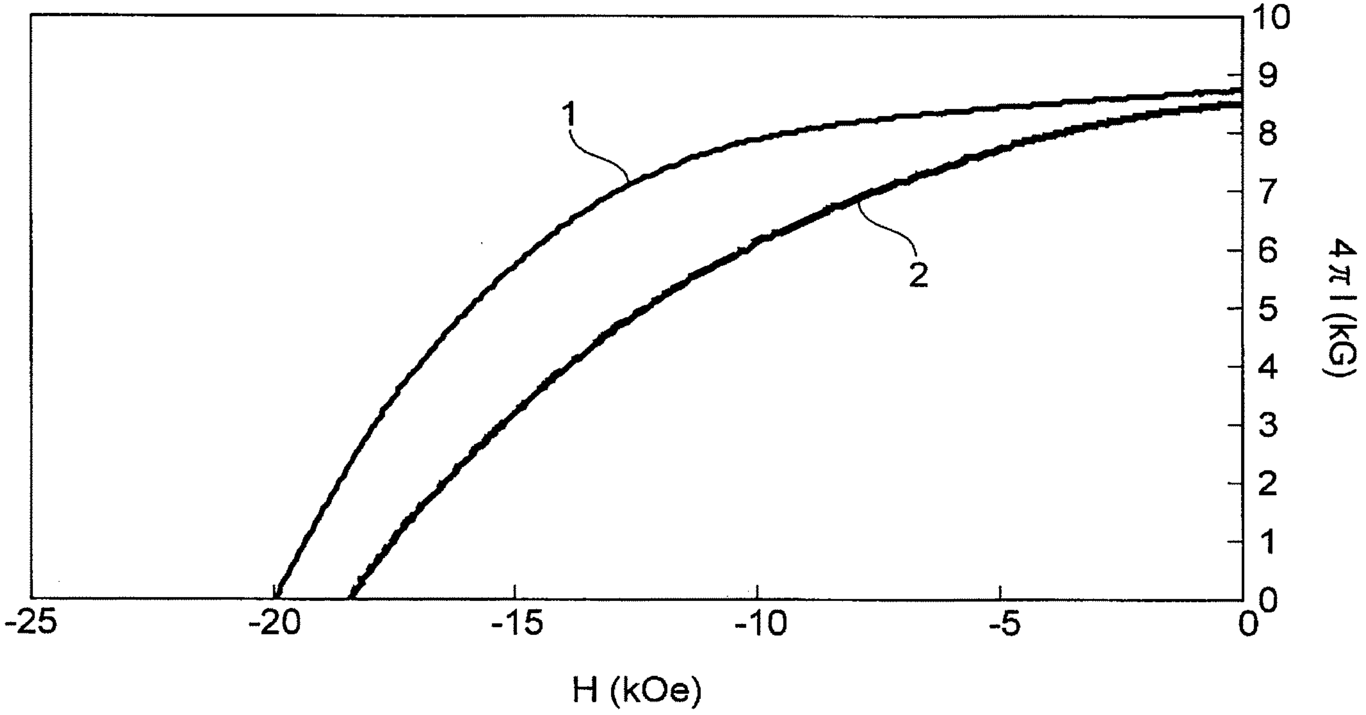


Fig.2



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EUROPEAN SEARCH REPORT

Application Number
EP 10 15 8135

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
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TECHNICAL FIELDS SEARCHED (IPC)

H01F
B22F
C22C

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
The Hague	31 May 2010	Straub, Florian

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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