

## Description

This invention relates to a process for producing a bulk-like permanent magnet suitable for use in a compact motor with a high output power, and more particularly, it relates to a process for producing a bulk-like permanent magnet directly from a melt spun powder of a rare earth-iron-boron material. The resulting bulk-like permanent magnet has an excellent demagnetizing force which is resistant to a strong demagnetizing field derived from an armature reaction. The bulk-like permanent magnet also has a high coercive force and a high residual induction, which are concerned with an improvement in the output power of motors. According to the process of this invention, bulk-like permanent magnets having such excellent characteristics can be produced with high dimensional precision and high productivity.

A permanent magnetic material in the non-equilibrium state or a metastable permanent magnetic material can be obtained by rapid solidification of a rare earth-iron-boron material with a melt spinning technique to solidify at least one part of the melted alloy without causing its crystallization. It is known that the resulting permanent magnetic material has a high coercive force and a high residual induction due to its non-equilibrium or metastable state (JP-A-59-64739). However, because the permanent magnetic material obtained by such a melt spinning technique is a powder in the form of thin ribbon or flake, it must be fixed to form a bulk-like permanent magnet suitable for use in a motor.

Examples of methods for fixing a melt spun powder include powder metallurgy methods such as non-pressure sintering processes. However, when a melt spun powder of a rare earth-iron-boron material is sintered without applying pressure, the excellent magnetic characteristics based on the non-equilibrium or metastable state may be degraded.

To solve this problem, a method for fixing a melt spun powder by plastic deformation has been proposed. This method comprises the steps of: charging a melt spun powder of a rare earth-iron-boron material into the cavity of a graphite mold; and fixing the melt spun powder by hot pressing with an induction heating system, thereby causing plastic deformation of the melt spun powder together with diffusion of atoms at the interface between the adhered powder particles, to form a bulk-like permanent magnet (JP-A-60-100402). The degree of fixation depends on the viscosity of the melt spun powder. When a melt spun powder having a low viscosity is used, a high degree of fixation can be obtained. However, it is necessary to heat the melt spun powder to a temperature higher than or equal to the crystallization temperature, for example, 600°C to 900°C, for the purpose of attaining a sufficient decrease in viscosity. Usually, several hours are required for heating the melt spun powder up to such a high temperature, after charging the powder into the cavity of a mold. The heating procedure for a long period of time may lead to a decrease in productivity. Also, because the melt spun powder reaches an equilibrium state, the excellent characteristics based on the non-equilibrium or metastable state may be degraded. Moreover, when the melt spun powder is simply compressed in the cavity of a mold, a high pressure of 1 to 3 ton/cm<sup>2</sup> (1 to 3 ton/cm<sup>2</sup>) must be applied in order to combine the powder particles with each other, because the surfaces of the powder particles do not have a low potential energy. Therefore, in this case, the durability of the mold will be decreased. In addition, the bulk-like permanent magnet prepared by the use of such a graphite mold does not have high dimensional precision. Therefore, the resulting bulk-like permanent magnet formed into nearly the desired shape must be further processed by grinding.

WO 89/12902 discloses a method for manufacturing a permanent magnet comprising generating Joule heat at contact interfaces between thin flakes of a rare earth-Fe alloy by applying uniaxial pressure to a gathered body of said thin flakes and supplying a current thereto, and bonding said gathered body integrally by making said thin flakes deform plastically in a worm state.

JP-A-1-175 705 discloses a method for producing a rare earth magnet comprising applying an arc discharge to an alloy powder and carrying out compression molding.

The process of this invention for producing a rare earth-iron-boron magnet comprises the steps of: charging a melt spun powder of a rare earth-iron-boron material into at least one cavity, wherein the cavity is formed between a pair of electrodes which are inserted into a hole in an electrically non-conductive ceramic die; subjecting the melt spun powder to a non-equilibrium plasma treatment, while applying a uniaxial pressure 20 MPa to 50 MPa (200 to 500 kgf/cm<sup>2</sup>) to the melt spun powder in the direction connecting the electrodes interposed between a pair of heat-compensating members under a reduced atmosphere of 100 to 1 Pa (10<sup>-1</sup> to 10<sup>-3</sup> Torr), thereby causing the fixation of the melt spun powder; and heating the fixed melt spun powder to a temperature higher than or equal to the crystallization temperature thereof by transferring, to the melt spun powder, Joule heat generated in the thermal compensating members when a current is allowed to pass through the members, thereby causing the plastic deformation of the melt spun powder to form a rare earth-iron-boron magnet; characterized in that the electrodes have a  $\rho/s.c$  value in the order of 10<sup>-5</sup>-10<sup>-4</sup>, and the thermal compensating members have a  $\rho/s.c$  value in the order of 10<sup>-3</sup>, where  $\rho$  is the specific resistance,  $s$  the specific gravity, and  $c$  the specific heat.

When electrodes and thermal compensating members having the  $p/s.c$  values are used, it is possible to heat the melt spun powder more uniformly. This is because when the amount of current flowing through the electrodes varies, the Joule heat generated in the thermal compensating members can be transferred uniformly to the melt spun powder.

In a preferred embodiment, a plurality of the electrically non-conductive ceramic dies having at least one pair of electrodes are stacked up on each other in the direction of applying the uniaxial pressure with each of the ceramic dies placed between a pair of thermal compensating members. If a mold having such a constitution is employed, it is possible to raise productivity.

In a preferred embodiment, the aforementioned rare earth-iron-boron material contains 13% to 15% of rare earth elements including yttrium (Y), 0% to 20% of cobalt (Co), 4% to 11% of boron (B), and the balance of iron (Fe) and impurities.

Thus, the invention described herein seeks to provide (1) a process for producing a rare earth-iron-boron magnet, by which a plurality of bulk-like permanent magnets can be prepared directly from a melt spun powder of a rare earth-iron-boron material; (2) a process for producing a rare earth-iron-boron magnet, in which the resulting bulk-like permanent magnet is magnetically isotropic, but has a lower residual induction than that of a permanent magnet prepared by non-pressure sintering, so that it is suitable for radial-directional magnetization; (3) a process for producing a rare earth-iron-boron magnet, which does not require subsequent processing of the resulting bulk-like permanent magnet by grinding, thereby increasing productivity; (4) a process for producing a rare earth-iron-boron magnet which can provide a bulk-like permanent magnet without degrading the excellent characteristics of a melt spun powder based on the non-equilibrium or metastable state; (5) a process for producing a rare earth-iron-boron magnet, which can simultaneously provide a plurality of bulk-like permanent magnets having a density close to the theoretical value, thereby attaining the same productivity as processes for producing resin bonded magnets; and (6) a process for producing a rare earth-iron-boron magnet, which can provide a bulk-like permanent magnet having quite excellent magnetic characteristics as compared with resin bonded magnets.

This invention may be better understood by reference to the accompanying drawing as follows:

Figure 1 is a partially-cutaway perspective view showing a mold used in the process for producing a rare earth-iron-boron magnet of this invention.

In the process of this invention, a bulk-like permanent magnet is prepared directly from a melt spun powder of a rare earth-iron-boron material. The rare earth-iron-boron material which can be used in the process of this invention preferably contains 13% to 15% of rare earth elements including yttrium (Y), 0% to 20% of cobalt (Co), 4% to 11% of boron (B), and the balance of iron (Fe) and impurities. Examples of the rare earth elements other than yttrium include neodymium (Nd) and praseodymium (Pr), which can provide a melt spun powder having a high coercive force.

When the amount of rare earth elements is less than 13%, the resulting melt spun powder will have not only a low coercive force but also a high deformation resistance. Thus, a bulk-like permanent magnet with a high induction cannot be obtained from such a melt spun powder. On the other hand, when the amount of rare earth elements is more than 15%, the melt spun powder will have a reduced saturation magnetization. Also, when pressure is applied to the melt spun powder in the process of this invention, because an excess amount of rare earth elements causes the formation of flashes or fins, the operation for producing a bulk-like permanent magnet will become difficult.

Although the inclusion of cobalt instead of a certain amount of iron increases the Curie point of the melt spun powder, when more than 20% of cobalt is added, a melt spun powder having a high coercive force cannot be obtained.

The amount of boron is preferably 4% to 11% in order to obtain the excellent magnetic characteristics derived from the  $R_2TM_{14}B$  phase present in the melt spun powder, wherein R is a rare earth element including yttrium, and TM is iron and/or cobalt. More preferably, the amount of boron is about 6% because it is then possible to obtain a melt spun powder with the minimum plastic deformation resistance.

The following is a description of a mold used in the process of this invention by reference to the accompanying figure.

Figure 1 shows a mold used in the process of this invention. With the use of this mold, a plurality of bulk-like permanent magnets with high dimensional precision can be prepared directly from a melt spun powder without losing the excellent magnetic characteristics based on the non-equilibrium or metastable state. The mold is comprised of an electrically non-conductive ceramic die 1 having at least one through hole  $1_{1-n}$ , at least one pair of electrodes  $2a_{1-n}$  and  $2b_{1-n}$ , and a pair of thermal compensating members 3a and 3b. The electrodes  $2a_{1-n}$  and  $2b_{1-n}$  are inserted into the through holes  $1_{1-n}$  to form cavities. These electrodes also function as upper and lower punches. The surfaces of the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  forming cavities are desirably coated with a layer containing boron nitrate powder. The electrically non-conductive ceramic die 1 having the electrodes

2a<sub>1-n</sub> and 2b<sub>1-n</sub> is placed between two thermal compensating members 3a and 3b. A melt spun powder 4<sub>1-n</sub> which is to be formed into a bulk-like permanent magnet is charged into the cavities.

The following is a description of the process of this invention using the above-mentioned mold.

First, the melt spun powder 4<sub>1-n</sub> is charged into the cavities between at least one pair of electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub>. After the electrically non-conductive ceramic die 1 having the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub> is placed between two thermal compensating members 3a and 3b, a uniaxial pressure of 20 MPa to 50 MPa (200 to 500 kgf/cm<sup>2</sup>) per cross area of the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub> in the direction connecting the electrodes is applied under a reduced atmosphere of 100 to 1 Pa (10<sup>-1</sup> to 10<sup>-3</sup> Torr), thereby reducing the surface potential energy of the melt spun powder 4<sub>1-n</sub>.

Then, the melt spun powder 4<sub>1-n</sub> is subjected to a non-equilibrium plasma treatment. A non-equilibrium plasma is a plasma with a much lower gas temperature than the electron temperature. The plasma is generated by applying a DC voltage between the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub> under a reduced atmosphere of 100 to 1 Pa (10<sup>-1</sup> to 10<sup>-3</sup> Torr). The electrolytic gas present in the plasma contains a large number of active atoms, molecules, ions, free electrons, radicals, and the like. The electron temperature is increased to about 10<sup>4</sup>°C by acceleration of the electrons in an electric field, whereas the temperatures of the atomic species and molecular species which have relatively larger masses are increased to only about 100°C to 200°C. When a solid material is treated with the non-equilibrium plasma, its surface temperature depends on the temperatures of the atoms and molecules present in the plasma, i.e., its gas temperature. Therefore, the melt spun powder 4<sub>1-n</sub> which is being treated with the non-equilibrium plasma cannot reach the temperature of plastic deformation, or the temperature at which the atoms can be diffused on its surface. However, electrons, ions, excited species, and other active chemical species present in the plasma, which have a certain amount of kinetic energy, may collide with the surface of the melt spun powder 4<sub>1-n</sub>, so that these active chemical species react with contaminants and low molecular weight compounds adhered to the surface of the melt spun powder 4<sub>1-n</sub>, thereby causing further reduction of the potential energy of the melt spun powder 4<sub>1-n</sub>, which is called an etching effect.

After the melt spun powder 4<sub>1-n</sub> is treated with the non-equilibrium plasma as described above, a current is allowed to pass through the powder 4<sub>1-n</sub> by way of the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub> from the side faces of the thermal compensating members 3a and 3b, under a reduced atmosphere and pressure, thereby causing generation of Joule heat in the thermal compensating members 3a and 3b. The Joule heat is then transferred to the melt spun powder 4<sub>1-n</sub>. The rate of temperature increase  $\Delta T/\Delta t$  (°C/sec) in the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub>, and in the melt spun powder 4<sub>1-n</sub>, is determined by the formula:

$$\Delta T/\Delta t = \frac{0.2389 \times I^2 R}{C}$$

$$= \frac{0.2389 \times I^2 l \rho / \pi r^2}{\pi r^2 l s c}$$

$$= \frac{0.2389 \times I^2 \rho}{\pi^2 r^4 s c}$$

where I is the current value (A), R is the electric resistance ( $\Omega$ ), C is the heat capacity (cal/°C), c is the specific heat (cal/°C-g), s is the specific gravity,  $\rho$  is the specific resistance ( $\Omega$ -cm), l is the length (cm) along the direction of applying uniaxial pressure, and r is the diameter (cm) of a cross section perpendicular to the direction of applying uniaxial pressure.

As seen from the above formula, the rate of temperature increase  $\Delta T/\Delta t$  equals  $(\Delta i)^2 \rho/s-c$ , where  $\Delta i$  is the current density (A/cm<sup>2</sup>). Thus, it can be seen that the rate of temperature increase  $\Delta T/\Delta t$  is independent of the length l (cm), but proportional to a square of the current density  $\Delta i$  (A/cm<sup>2</sup>) as well as to the specific resistance  $\rho$  ( $\Omega$ -cm), and inversely proportional to the specific heat c (cal/°C-g) and the specific gravity s.

The melt spun powder 4<sub>1-n</sub> has a  $\rho/s-c$  value in the order of  $2.7 \times 10^{-4}$  at the initial stage. The electrodes

2a<sub>1-n</sub> and 2b<sub>1-n</sub> preferably have a slightly lower p/s-c value in the order of  $2.7 \times 10^{-4}$  or  $10^{-5}$ , and the thermal compensating members 3a and 3b have a p/s-c value in the order of  $10^{-3}$ . When a current is allowed to pass through the melt spun powder 4<sub>1-n</sub>, it does not necessarily flow uniformly because of the contact resistance in the electrodes. Therefore, the melt spun powder 4<sub>1-n</sub> does not have a constant rate of temperature increase. However, when the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub>, and the thermal compensating members 3a and 3b having the aforementioned ranges of p/s-c values are used, the Joule heat transferred is corrected, thereby providing the melt spun powder 4<sub>1-n</sub> with a constant rate of temperature increase.

The rate of temperature increase of the melt spun powder 4<sub>1-n</sub> depends mainly on the Joule heat generated in the thermal compensating members 3a and 3b when a current is applied. The melt spun powder 4<sub>1-n</sub> is heated to a temperature higher than the crystallization temperature thereof by transferring Joule heat, thereby causing plastic deformation at a strain rate of  $10^{-1}$  to  $10^{-2}$  mm/sec or more. The strain rate of the melt spun powder 4<sub>1-n</sub> increases with a decrease in the viscosity thereof and with an increase in the relative density thereof; it reaches a peak level and then gradually decreases. When the relative density of the melt spun powder 4<sub>1-n</sub> is more than 90%, the strain rate is already lower than its peak level. However, the current is applied until the strain rate reaches  $10^{-3}$  mm/sec or less. Although the current is shut off at the time that the strain rate becomes  $10^{-3}$  mm/sec or less, the pressure and reduced atmosphere are still maintained until the outer surface temperature of the non-conductive ceramic die 1 decreases. Thus, rare earth-iron-boron magnets having excellent magnetic characteristics based on the non-equilibrium or metastable state, as well as densification, can be obtained as bulk-like permanent magnets. With the use of a mold as shown in Figure 1, n bulk-like permanent magnets are prepared at a time, thereby attaining high productivity.

The resulting rare earth-iron-boron magnets are released from the non-conductive ceramic die 1 by use of a difference in the thermal expansion therebetween when cooled in the cavities. If the surfaces of the electrodes 2a<sub>1-n</sub> and 2b<sub>1-n</sub> which forms a cavity are coated with a layer containing boron nitride powder (i.e., releasing film), the magnets can also be released readily, because the boron nitride powder is transferred to the surface of the magnets.

The melt spun powder of a rare earth-iron-boron material which can be used in this invention is prepared by a well-known rapid solidification technique such as a melt spinning technique. The particle size of the melt spun powder is not particularly limited, but the amount of fine melt spun powder having a particle size of 53  $\mu$ m or less is preferably small, because this provides a rare earth-iron-boron magnet having a low coercive force.

Examples of the materials used for the electrodes include a hard metal alloy G5 defined by the specification of JIS H5501. Examples of the materials used for the thermal compensating members include graphite and various ceramic composites obtained by adding to SiC, about 30% to 50% by volume of at least one compound selected from the group consisting of TiC, TiN, ZnC, WC, ZrB<sub>2</sub>, HfB<sub>2</sub>, NbB<sub>2</sub> and TaB<sub>2</sub>, and sintering the mixture. Since the electrically non-conductive ceramic die has a small coefficient of thermal conductivity, it provides high thermal efficiency by the prevention of current and heat leaks. Also, the electrically non-conductive ceramic die is required to have excellent properties such as thermal shock resistance, inactivity to the melt spinning powder, wear resistance, a low thermal expansion coefficient, strength at high temperatures, and a low heat capacity. Examples of the materials used for the electrically non-conductive ceramic die include sialon which is a composite of silicon nitride and alumina.

The invention will be further illustrated by reference to the following examples, but these examples are not intended to restrict the invention.

#### Example 1

First, a rare earth-iron-boron material containing 13% of Nb, 68% of Fe, 18% of Co, and 6% of B was melted by high-frequency heating under an atmosphere of argon gas, and then sprayed onto a copper single roller at a peripheral velocity of about 50 m/sec by a melt spinning technique to obtain a melt spun powder in the form of a flake having a thickness of 20 to 30  $\mu$ m. It was confirmed by X-ray diffraction that the melt spun powder was formed by solidifying the melted alloy without causing its crystallization.

The melt spun powder in the non-equilibrium state was then ground to a particle size range between 53  $\mu$ m and 350  $\mu$ m. A part of the powder having the adjusted particle size was magnetized with a pulsed magnetic field of 50 kOe. The intrinsic coercive force of the melt spun powder thus magnetized was measured to be 5.8 kOe with a vibrating sample magnetometer (VSM).

On the other hand, a part of the melt spun powder having the adjusted particle size in the non-equilibrium state was heat-treated at a temperature of 650°C to 700°C under an atmosphere of argon gas. The presence of a R<sub>2</sub>Fe<sub>14</sub>B phase in the heat-treated powder was confirmed by X-ray diffraction. The powder was then magnetized with a pulsed magnetic field of 50 kOe, as described above. The intrinsic coercive force of the powder

thus magnetized was measured to be 16.5 kOe with a vibrating sample magnetometer (VSM). The resulting melt spun powder is referred to as a metastable rapid solidification powder in contrast with the melt spun powder in the non-equilibrium state.

Appropriate amounts of the powder in the non-equilibrium state and the metastable powder were independently weighed and charged into the cavities between the electrodes  $2a_{1-n}$  and  $2b_{1-n}$ , as shown in Figure 1. The electrically non-conductive ceramic die 1 had through holes  $1_{1-n}$  having a diameter of 14 mm. The electrodes  $2a_{1-n}$  and  $2b_{1-n}$  were inserted into the respective through holes  $1_{1-n}$  to form the cavities. Also, the electrically non-conductive ceramic die 1, and the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  forming the cavities were placed between the two thermal compensating members 3a and 3b. A plurality of bulk-like permanent magnets were prepared from the melt spun powder  $4_{1-n}$  which had been charged into the cavities according to the following procedure.

In this example, the subscript "n" was 10, and therefore, ten cavities were formed by inserting the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  into the through holes  $1_{1-n}$ . The electrodes  $2a_{1-n}$  and  $2b_{1-n}$  also functioned as upper and lower punches, respectively. The electrodes  $2a_{1-n}$  and  $2b_{1-n}$  were made of a hard metal alloy G5 defined by the specification of JIS H5501, or a SiC/TiC ceramic composite containing a certain amount of TiC. The surface of the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  forming the cavities had been previously coated with a layer containing boron nitride powder. Also, the electrically non-conductive ceramic die was made of sialon. The thermal compensating members 3a and 3b were made of graphite or an SiC/TiC ceramic composite containing a certain amount of TiC.

Next, a uniaxial pressure of 20 MPa to 50 MPa (200 to 500 kgf/cm<sup>2</sup>) per cross-sectional area of the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  perpendicular to the direction connecting the electrodes was applied to the powder  $4_{1-n}$  under a reduced atmosphere of 100 to 1 Pa ( $10^{-1}$  to  $10^{-3}$  Torr). Then, the powder  $4_{1-n}$  was subjected to a non-equilibrium plasma treatment by applying a DC voltage of 10 V having a pulse length of 20 msec between the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  for zero to 90 seconds, while keeping the reduced atmosphere and pressure constant. Subsequently, a DC current of 300 to 350 A/cm<sup>2</sup> per cross-sectional area of the electrodes  $2a_{1-n}$  and  $2b_{1-n}$  perpendicular to the direction connecting the electrodes was allowed to pass through the powder  $4_{1-n}$  by way of these electrodes from the sides of the thermal compensating members 3a and 3b for 40 to 500 seconds. At that time, the powder  $4_{1-n}$  present in the cavities was heated and compressed in the direction of applying the pressure. The strain rate was determined by obtaining the value of displacement of the powder  $4_{1-n}$  thus heated, and then differentiating the value. The viscosity of the powder  $4_{1-n}$  was rapidly reduced by heating and application of a constant pressure, whereas the strain rate was increased. However, when the relative density of the powder  $4_{1-n}$  exceeded 90%, the strain rate started decreasing with an increase in the relative density. The current was shut off when the strain rate became  $10^{-3}$  mm/sec or less. When the outer surface temperature of the electrically non-conductive ceramic die 1 started decreasing, the pressure and the reduced atmosphere were released. In this way, ten bulk-like permanent magnets having a diameter of 14 mm and a height of 2 mm were obtained directly from a melt spun powder of a rare earth-iron-boron material.

Table 1 shows the relationship between the non-equilibrium plasma treatment time and the intrinsic coercive force of the bulk-like permanent magnets prepared from either the melt spun powder in the non-equilibrium state or the metastable melt spun powder in the case where the electrodes had a  $\rho/s-c$  value in the order of  $10^{-5}$ , and the thermal compensating members had a  $\rho/s-c$  value in the order of  $10^{-3}$ , where  $\rho$  is the specific resistance ( $\Omega\cdot\text{cm}$ ),  $s$  is the specific gravity, and  $c$  is the specific heat (cal/°C.g). As can be seen from the Table, a bulk-like permanent magnet having an intrinsic coercive force of 15 kOe or more can be obtained from either the melt spun powder in the non-equilibrium or the metastable melt spun powder by a non-equilibrium plasma treatment.

Table 1

| Non-equilibrium plasma        |     |      |      |      |
|-------------------------------|-----|------|------|------|
| treatment time (sec)          |     |      |      |      |
|                               | 0   | 30   | 60   | 90   |
| Intrinsic coercive force of a |     |      |      |      |
| bulk-like permanent magnet    |     |      |      |      |
| obtained from melt spun       |     |      |      |      |
| powder in the non-equilibrium |     |      |      |      |
| state (kOe)                   |     |      |      |      |
|                               | 8.8 | 16.8 | 17.2 | 17.4 |
| Intrinsic coercive force of a |     |      |      |      |
| bulk-like permanent magnet    |     |      |      |      |
| obtained from metastable      |     |      |      |      |
| melt spun powder (kOe)        |     |      |      |      |
|                               | 7.5 | 13.7 | 16.6 | 17.0 |

Table 2 shows the relationship between current-applying time, and the intrinsic coercive force and residual induction of the bulk-like permanent magnet in the case where the electrodes had a  $\rho/s-c$  value in the order of  $10^{-3}$  to  $10^{-5}$ , and the thermal compensating members had a  $\rho/s-c$  value in the order of  $10^{-3}$  to  $10^{-4}$ , where  $\rho$  is the specific resistance ( $\Omega\text{-cm}$ ),  $s$  is the specific gravity, and  $c$  is the specific heat ( $\text{cal}/^\circ\text{C}\cdot\text{g}$ ). As can be seen from the Table, a bulk-like permanent magnet having stable magnetic properties can be obtained when electrodes having a  $\rho/s-c$  value in the order of  $10^{-4}$ , and thermal compensating members having a  $\rho/s-c$  value in the order of  $10^{-3}$  are used with a relatively short current-applying time according to the method of this invention.

Table 2

|                                                                           | Ex. 1       | Comp.<br>Ex. 1 | Comp.<br>Ex. 2 | Comp.<br>Ex. 3 |
|---------------------------------------------------------------------------|-------------|----------------|----------------|----------------|
| $\rho/s \cdot c$ value of<br>thermal compen-<br>sating members            | $10^{-3}$   | $10^{-3}$      | $10^{-4}$      | $10^{-4}$      |
| $\rho/s \cdot c$ value of<br>electrodes                                   | $10^{-4}$   | $10^{-3}$      | $10^{-4}$      | $10^{-3}$      |
| Current-<br>applying<br>time (sec)                                        | 70-80       | 30-60          | 450-500        | 50-70          |
| Intrinsic coer-<br>cive force of<br>bulk-like perma-<br>nent magnet (kOe) | 16-17       | 11-14          | 9-14           | 9-17           |
| Residual induction<br>of bulk-like<br>permanent<br>magnet (kG)            | 8.3-<br>8.4 | 7.9-<br>8.0    | 7.8-<br>8.0    | 7.9-<br>8.2    |

When electrodes having a  $\rho/s \cdot c$  value in the order of  $10^{-4}$  and thermal compensating members having a  $\rho/s \cdot c$  value in the order of  $10^{-3}$  were used as described in Table 2, a bulk-like permanent magnet having an outer diameter of  $14.000 \pm 0.01$  mm, a height of  $2.00 \pm 0.05$  mm, and a density of 7.68 to 7.70 g/cm<sup>3</sup>, was obtained.

#### Example 2

Twenty bulk-like permanent magnets were prepared in the same manner as that of Example 1, except that two molds as shown in Figure 1 were stacked up on each other in the direction of applying uniaxial pressure with each of the electrically non-conductive ceramic dies placed between a pair of thermal compensating members. The bulk-like permanent magnets obtained by applying a current for the same period of time as that of Example 1, had substantially the same magnetic properties, dimensional precision, and density as those of Example 1.

#### Claims

1. A process for producing a rare earth-iron-boron magnet comprising the steps of:

**Table 2**

|                                                                                     | <b>Ex. 1</b> | <b>Comp.<br/>Ex. 1</b> | <b>Comp.<br/>Ex. 2</b> | <b>Comp.<br/>Ex. 3</b> |
|-------------------------------------------------------------------------------------|--------------|------------------------|------------------------|------------------------|
| <b><math>\rho/s \cdot c</math> value of<br/>thermal compen-<br/>sating members</b>  | $10^{-3}$    | $10^{-3}$              | $10^{-4}$              | $10^{-4}$              |
| <b><math>\rho/s \cdot c</math> value of<br/>electrodes</b>                          | $10^{-4}$    | $10^{-3}$              | $10^{-4}$              | $10^{-3}$              |
| <b>Current-<br/>applying<br/>time (sec)</b>                                         | 70-80        | 30-60                  | 450-500                | 50-70                  |
| <b>Intrinsic coer-<br/>cive force of<br/>bulk-like perma-<br/>nent magnet (kOe)</b> | 16-17        | 11-14                  | 9-14                   | 9-17                   |
| <b>Residual induction<br/>of bulk-like<br/>permanent<br/>magnet (kG)</b>            | 8.3-<br>8.4  | 7.9-<br>8.0            | 7.8-<br>8.0            | 7.9-<br>8.2            |

When electrodes having a  $\rho/s \cdot c$  value in the order of  $10^{-4}$  and thermal compensating members having a  $\rho/s \cdot c$  value in the order of  $10^{-3}$  were used as described in Table 2, a bulk-like permanent magnet having an outer diameter of  $14.000 \pm 0.01$  mm, a height of  $2.00 \pm 0.05$  mm, and a density of 7.68 to 7.70 g/cm<sup>3</sup>, was obtained.

#### Example 2

Twenty bulk-like permanent magnets were prepared in the same manner as that of Example 1, except that two molds as shown in Figure 1 were stacked up on each other in the direction of applying uniaxial pressure with each of the electrically non-conductive ceramic dies placed between a pair of thermal compensating members. The bulk-like permanent magnets obtained by applying a current for the same period of time as that of Example 1, had substantially the same magnetic properties, dimensional precision, and density as those of Example 1.

#### **Claims**

1. A process for producing a rare earth-iron-boron magnet comprising the steps of:



charging a melt spun powder of a rare earth-iron-boron material into at least one cavity, wherein said cavity is formed between a pair of electrodes ( $2a_{1-n}$ ,  $2b_{1-n}$ ) which are inserted into a through hole ( $1_{1-n}$ ) in an electrically non-conductive ceramic die (1);

subjecting said melt spun powder to a non-equilibrium plasma treatment, while applying a uniaxial pressure of 20 MPa to 50 MPa (200 to 500 kgf/cm<sup>2</sup>) to said melt spun powder in the direction connecting said electrodes interposed between a pair of heat-compensating members (3a, 3b) under a reduced atmosphere of 100 to 1 Pa ( $10^{-1}$  to  $10^{-3}$  Torr), thereby causing the fixation of said melt spun powder; and

heating the fixed melt spun powder to a temperature higher than or equal to the crystallization temperature thereof by transferring, to said melt spun powder, Joule heat generated in said thermal compensating members (3a, 3b) when a current is allowed to pass through said members thereby causing the plastic deformation of said melt spun powder to form a rare earth-iron boron magnet;

characterised in that said electrodes have a  $\rho/s \cdot c$  value in the order of  $10^{-6}$ - $10^{-4}$ , and said thermal compensating members have a  $\rho/s \cdot c$  value in the order of  $10^{-3}$ , where  $\rho$  is the specific resistance,  $s$  the specific gravity, and  $c$  the specific heat.

2. A process according to claim 1, wherein a plurality of said electrically non-conductive ceramic dies (1) having at least one pair of electrodes (2a, 2b) are stacked up on each other in the direction of applying said uniaxial pressure with each of said ceramic dies placed between a pair of thermal compensating members (3a, 3b).

3. A process according to claim 1 or 2, wherein said rare earth-iron-boron material (4) comprises 13% to 15% of rare earth elements including yttrium (Y), 0% to 20% of cobalt (Co), 4% to 11% of boron (B), and the balance of iron (Fe) and impurities.

## Patentansprüche

1. Verfahren zur Herstellung eines Seltenerd-Eisen-Bor Magneten, bei dem man ein schmelzgesponnenes Pulver eines Seltenerd-Eisen-Bor Materials in mindestens eine Höhlung füllt, welche zwischen einem Paar Elektroden ( $2a_{1-n}$ ,  $2b_{1-n}$ ) ausgebildet ist, die durch ein Loch ( $1_{1-n}$ ) in einer elektrisch nicht leitfähigen Keramikform (1) eingeführt werden, dieses schmelzgesponnenen Pulver einer Ungleichgewichtsplasmabehandlung unterzieht, während ein uniaxialer Druck von 20 MPa bis 50 MPa (200 bis 500 kgf/cm<sup>2</sup>) in der Richtung aufgebracht wird, welche die zwischen ein Paar wärmekompensierende Komponenten (3a, 3b) gelegten Elektroden bei verringerter Atmosphäre von 100 bis 1 Pa ( $10^{-1}$  bis  $10^{-3}$  Torr) verbindet und dadurch die Fixierung des schmelzgesponnenen Pulvers bewirkt, und das fixierte schmelzgesponnene Pulver auf eine Temperatur erhitzt, die höher als seine Kristallisationstemperatur oder gleich derselben ist, indem man auf das schmelzgesponnene Pulver Joule-Wärme überträgt, die in den wärmekompensierenden Komponenten (3a, 3b) erzeugt wird, wenn man einen Strom durch diese Komponenten leitet, wodurch die plastische Deformation des schmelzgesponnenen Pulvers bewirkt und ein Seltenerd-Eisen-Bor Magnet erzeugt wird; dadurch gekennzeichnet, daß die Elektroden einen  $\rho/s \cdot c$ -Wert in der Größenordnung von  $10^{-6}$  bis  $10^{-4}$  und die wärmekompensierenden Komponenten einen  $\rho/s \cdot c$  Wert in der Größenordnung von  $10^{-3}$  aufweisen, wobei  $\rho$  der spezifische Widerstand,  $s$  das spezifische Gewicht und  $c$  die spezifische Wärme ist.

2. Verfahren nach Anspruch 1; bei dem eine Vielzahl dieser elektrisch nicht leitfähigen Keramikformen (1) mit mindestens einem Elektrodenpaar (2a, 2b) in der Richtung aufeinandergestapelt werden, in der der uniaxiale Druck aufgebracht wird, wobei jede der Keramikformen zwischen ein Paar wärmekompensierende Komponenten (3a, 3b) gelegt wird.

3. Verfahren nach Anspruch 1 oder 2, bei dem das Seltenerd-Eisen-Bor Material (4) 13 % bis 15 % Seltenerdmetalle, darunter Yttrium (Y), 0 % bis 20 % Cobalt (Co) und 4 % bis 11 % Bor (B) umfaßt und der Rest aus Eisen (Fe) und Verunreinigungen besteht.

## Revendications

1. Procédé de production d'un aimant à base de terre rare-fer-bore, comprenant les étapes consistant à:



charger une poudre centrifugée en fusion d'un matériau de terre rare-fer-bore dans au moins une cavité, où ladite cavité est formée entre une paire d'électrodes ( $2a_{1-n}$ ,  $2b_{1-n}$ ) qui sont insérées dans un trou traversant ( $1_{1-n}$ ) dans un moule céramique électriquement non-conducteur (1),

5 soumettre ladite poudre centrifugée en fusion à un traitement au plasma à l'état de non-équilibre tout en appliquant une pression uniaxiale de 20 MPa à 50 MPa (200 à 500 kgf/cm<sup>2</sup>) sur cette poudre centrifugée en fusion dans la direction reliant lesdites électrodes interposées entre une paire d'éléments compensateurs de chaleur (3a, 3b) sous une atmosphère réduite de 100 à 1 Pa ( $10^{-1}$  à  $10^{-3}$  Torr), provoquant ainsi la fixation de ladite poudre centrifugée en fusion, et

10 chauffer la poudre centrifugée en fusion à une température supérieure ou égale à sa température de cristallisation en transférant à ladite poudre centrifugée en fusion de la chaleur Joule générée dans lesdits éléments compensateurs thermiques (3a, 3b) lorsqu'un courant peut passer dans lesdits éléments, provoquant ainsi la déformation plastique de ladite poudre centrifugée en fusion pour former un aimant à base de terre rare-fer-bore,

15 caractérisé en ce que lesdites électrodes ont une valeur  $p/s.c$  de l'ordre de  $10^{-6}$  à  $10^{-4}$ , et que lesdits éléments compensateurs thermiques ont une valeur  $p/s.c$  de l'ordre de  $10^{-3}$ , où  $p$  est la résistance spécifique,  $s$  est la gravité spécifique et  $c$  est la chaleur spécifique.

2. Procédé suivant la revendication 1, dans lequel une pluralité desdits moules céramiques électriquement non-conducteurs (1) présentant au moins une paire d'électrodes (2a, 2b) sont empilés l'un sur l'autre dans la direction de l'application de ladite pression uniaxiale, chacun desdits moules céramiques étant placé entre une paire d'éléments compensateurs thermiques (3a, 3b).

3. Procédé suivant la revendication 1 ou 2, dans lequel ledit matériau de terre rare-fer-bore (4) comprend de 13% à 15% d'éléments de terres rares, y compris yttrium (Y), de 0% à 20% de cobalt (Co), de 4% à 11% de bore (B) et, pour le reste, du fer (Fe) et des impuretés.