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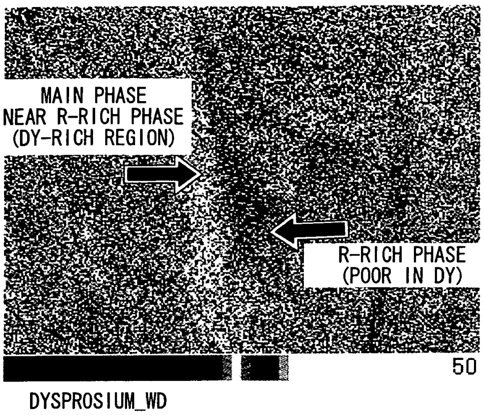
(54) R-T-B ALLOY, METHOD FOR PRODUCING THE SAME, FINE POWDER FOR R-T-B RARE EARTH PERMANENT MAGNET, AND R-T-B RARE EARTH PERMANENT MAGNET

(57) An object of the present invention is to provide an R-T-B type alloy (wherein R is at least one element selected from Sc, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb, and Lu; T is a transition metal that contains 80% by mass or more of Fe; and B is one that contains 50% by mass or more of boron (B) and also contains at least one element of C and N within a range from 0 to less than 50% by mass) that contains at least Dy, as a raw material for a rare earth-based permanent magnet having excellent magnetic characteristics, and the R-T-B type alloy provided in the present invention includes a main phase such as an $R_2T_{14}B$ phase for exhibiting magnetic properties; an R-rich phase that is relatively enriched with R compared to the overall alloy compositional ratio; and a Dy-rich region that is formed close to the R-rich phase and relatively enriched with Dy compared to the compositional ratio.

FIG. 8



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Description

TECHNICAL FIELD

[0001] The present invention relates to an R-T-B type alloy, a fine powder for an R-T-B type rare earth permanent magnet, and an R-T-B type rare earth permanent magnet. In particular, the present invention relates to an R-T-B type alloy and a fine powder for an R-T-B type rare earth permanent magnet that can provide an R-T-B type rare earth permanent magnet having excellent coercive force.

BACKGROUND ART

[0002] R-T-B type magnets have been used for hard disks (HD), magnetic resonance imaging (MRI), various types of motors and the like for their high-performance characteristics. A recent increase in demand for energy saving, in addition to enhancements in the heat resistance of R-T-B type magnets, has caused the usage rate in motors, including automobile motors, to increase.

R-T-B type magnets have Nd, Fe and B as the main components and thus, the magnets of this type are collectively called Nd-Fe-B type or R-T-B type magnets. In an R-T-B type magnet, R is primarily Nd with a part being replaced by another rare earth element such as Pr, Dy and Tb; T is Fe with a part being replaced by another transition metal such as Co and Ni; and B is boron and may be partially replaced by C or N.

[0003] The R-T-B type alloy which can be used in an R-T-B type magnet is an alloy where a magnetic $R_2T_{14}B$ phase contributing to the magnetization activity is the main phase and coexists with a non-magnetic, rare earth element-enriched and low-melting point R-rich phase. Since this R-T-B type alloy is an active metal, it is generally melted or cast in vacuum or in an inert gas. From the cast R-T-B type alloy ingot, a sintered magnet is usually produced by a powder metallurgy process as follows. The alloy ingot is ground into an alloy powder with an average particle size of about 5 μm (d50: measured by a laser-diffraction particle size distribution analyzer), press-shaped in a magnetic field, sintered at a high temperature of about 1,000 to 1,100°C in a sintering furnace, then subjected to, if necessary, heat treatment and machining, and further plated for enhancing the corrosion resistance, thereby completing a sintered magnet.

[0004] In the R-T-B type sintered magnet, the R-rich phase plays the following important roles:

1) forming a liquid phase during the sintering by virtue of a low melting point and thereby contributing to high densification of the magnet and in turn, enhancement of the magnetization;
2) eliminating unevenness on the grain boundary and thereby yielding a reduction in the nucleation site of the reversed magnetic domain and an increase in the coercive force; and
3) magnetically isolating the main phase and thereby increasing the coercive force.

Accordingly, if the R-rich phase in the shaped magnet is in a poorly dispersed state, it incurs local failure of sintering or reduction of magnetism. Therefore, it is important that the R-rich phase is uniformly dispersed in the shaped magnet. The R-rich phase distribution in the R-T-B type sintered magnet is greatly affected by the texture of the raw material R-T-B type alloy.

[0005] Another problem encountered in casting an R-T-B type alloy is the production of α-Fe in the cast alloy. The α-Fe has deformability and remains in the grinder without being ground, and this not only decreases the grinding efficiency at the grinding of the alloy but also affects the compositional fluctuation or particle size distribution before and after grinding. If α-Fe still remains in the magnet after sintering, a reduction in the magnetic characteristics of the magnet results. Accordingly, an alloy has been heretofore subjected to a homogenization treatment at a high temperature for a long period of time, where necessary, to eliminate α-Fe. However, α-Fe is present as a peritectic nucleus, and therefore its elimination requires solid phase diffusion for a long period of time. In the case of an ingot having a thickness of several centimeters and a rare earth content of 33% or less, elimination of α-Fe is practically impossible.

[0006] In order to solve the problem that α-Fe is produced in the R-T-B type alloy, a strip casting method (simply referred to as an "SC method") of casting an alloy ingot at a higher cooling rate has been developed and used. The SC method is a method of solidifying an alloy through rapid cooling, where a molten alloy is cast on a copper roll the inside of which is water-cooled, and a flake of about 0.1 to 1 mm is produced. In the SC method, the molten alloy is supercooled down to a temperature where the main $R_2T_{14}B$ phase is produced or even lower, so that an $R_2T_{14}B$ phase can be produced directly from a molten alloy and the formation of α-Fe can be suppressed. Furthermore, in the SC method, a fine microstructure is generated in the alloy, so that an alloy having a microstructure allowing for fine dispersion of an R-rich phase can be produced. The R-rich phase expands by reacting with hydrogen in a hydrogen atmosphere and becomes a brittle hydride. By utilizing this property, fine cracking commensurate with the dispersion degree of the R-rich phase can be introduced. When an alloy is pulverized through this hydrogenation step, a large number of fine cracks are produced by the hydrogenation trigger breakage of the alloy, and therefore very good grindability is attained. The internal R-rich phase in the alloy produced by the SC method is thus finely dispersed, and this leads to good dispersibility of the R-rich phase also in the magnet after grinding and sintering, thereby enhancing the magnetic characteristics of the magnet (see, for example, Patent Document 1).

[0007] The alloy flake produced by the SC method is also excellent in terms of microstructure homogeneity. The microstructure homogeneity can be compared by the crystal grain diameter or the dispersed state of the R-rich phase. In the case of an alloy flake produced by the SC method, a chill crystal is sometimes generated on the casting roll side of the alloy flake (hereinafter referred to as a "mold face side"), but an appropriately fine homogeneous texture yielded by the solidification through rapid cooling can be obtained as a whole. As described above, in the R-T-B type alloy produced by the SC method, the R-rich phase is finely dispersed and the formation of α-Fe is also suppressed, and thus the R-T-B type alloy has an excellent microstructure for the production of a sintered magnet.

[0008] The distribution of Dy contributing to enhancement of the coercive force greatly affects magnet characteristics, particularly the relationship between the coercive force and the element distribution in the microstructure of a magnet. For example, the coercive force has already been reported to be high when Dy is distributed close to the grain boundary phase (see, for example, Patent Document 2).

More specifically, the coercive force has also been reported to be high when Dy is present in the main phase (see, for example, Patent Document 3 and Non-patent Document 1).

Additionally, since there is a definite relationship between magnet characteristics and the alloy production methods, the methods for producing alloys have also advanced along with the improvements of magnet characteristics. For example, a method for controlling microstructures (see, for example, Patent Document 4) and a method for controlling microstructures by processing the surface of a casting roll to a predetermined roughness (see, for example, Patent Documents 5 and 6) are known.

[Patent Document 1] Japanese Unexamined Patent Application, First Publication No. Hei 5-222488
[Patent Document 2] Japanese Unexamined Patent Application, First Publication No. Hei 5-21219
[Patent Document 3] WO 2003/001541
[Patent Document 4] WO 2005/031023
[Patent Document 5] Japanese Unexamined Patent Application, First Publication No. 2003-188006
[Patent Document 6] Japanese Unexamined Patent Application, First Publication No. 2004-43291
[Non-Patent Document 1] Hiroyuki TOMIZAWA, Journal of the Japan Society of Powder and Powder Metallurgy, March, 2005, vol. 52, issue 3, pp. 158-163.

DISCLOSURE OF THE INVENTION

PROBLEMS TO BE SOLVED BY THE INVENTION

[0009] However, in recent years, R-T-B type rare earth permanent magnets with even higher performance have been required and thus, demands for further improvements in the magnetic characteristics, such as the coercive force, of R-T-B type rare earth permanent magnets are increasing.

The present invention has been made in view of the above circumstances and an object of the present invention is to provide an R-T-B type alloy as a raw material for a rare earth-based permanent magnet having excellent magnetic characteristics.

Another object of the present invention is to provide a fine powder for an R-T-B type rare earth permanent magnet produced from the above R-T-B type alloy, and an R-T-B type rare earth permanent magnet.

MEANS FOR SOLVING THE PROBLEMS

[0010] The present inventors have conducted a detailed observation of the texture of the R-T-B type alloy containing Dy to be used for producing R-T-B type rare earth permanent magnets to investigate the relationship between the texture state and the magnetic characteristics. Also, the present inventors have confirmed the fact that when the R-T-B type alloy containing Dy includes a Dy-rich region enriched with Dy in addition to the main phase formed from an $R_2T_{14}B$ phase and an R-rich phase enriched with R, the R-T-B type rare earth permanent magnet obtained by shaping/sintering a fine powder, which is produced from the flakes of this R-T-B type alloy, will have excellent magnetic characteristics such as coercive force. The present invention has been accomplished based on these findings.

[0011] That is, the present invention provides the following.

(1) An R-T-B type alloy (in which R is at least one element selected from Sc, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb, and Lu; T is a transition metal that contains 80% by mass or more of Fe; and B is one that contains 50% by mass or more of boron (B) and also contains at least one element of C and N within a range from

0 to less than 50% by mass) which is a raw material for use in a rare earth-based permanent magnet and contains at least Dy, comprising a main phase such as an $R_2T_{14}B$ phase for exhibiting magnetic properties, an R-rich phase that is relatively enriched with R compared to the overall alloy compositional ratio, and a Dy-rich region that is formed close to the R-rich phase and relatively enriched with Dy compared to the aforementioned compositional ratio.

(2) The R-T-B type alloy as described in (1), wherein Dy concentration is lower in the main phase than in the Dy-rich region and is lower in the R-rich phase than in the main phase.

(3) The R-T-B type alloy as described in (1) or (2), wherein the alloy is a flake having an average thickness from 0.1 to 1 mm produced by a strip casting method.

[0012]

(4) A method for producing an R-T-B type alloy described in any one of the above (1) to (3), comprising the steps of: producing a flake having an average thickness from 0.1 to 1 mm, and supplying molten alloy to a cooling roll at an average rate of 10 g/sec or more per 1-cm width.

(5) The method for producing an R-T-B type alloy described in (4), characterized in that an R-T-B type alloy flake rolled out of the cooling roll is kept at a temperature from 600 to 900°C for 30 seconds or more.

[0013]

(6) A fine powder for an R-T-B type rare earth permanent magnet that is produced from the R-T-B type alloy described in any one of the above (1) to (3) or from the R-T-B type alloy produced by the method for producing an R-T-B type alloy described in (4) or (5).

(7) An R-T-B type rare earth permanent magnet produced from the fine powder for an R-T-B type rare earth permanent magnet described in (6).

EFFECTS OF THE INVENTION

[0014] The R-T-B type alloy of the present invention is formed close to the R-rich phase and has a Dy-rich region relatively enriched with Dy compared to the overall compositional ratio. Accordingly, a rare earth permanent magnet having a high coercive force and excellent magnetic characteristics can be achieved.

Also, the fine powder for an R-T-B type rare earth permanent magnet and the R-T-B type rare earth permanent magnet of the present invention are produced from either the R-T-B type alloy of the present invention or the R-T-B type alloy produced by the method of the present invention for producing an R-T-B type alloy, and thus will have a high coercive force and excellent magnetic characteristics.

BRIEF DESCRIPTION OF THE DRAWINGS

[0015]

FIG. 1 is a photograph showing one example of the R-T-B type alloy of the present invention. The photograph is taken when the cross-section of the R-T-B type alloy flake is observed by a scanning electron microscope (SEM).

FIG. 2 is an electron image of the R-T-B type alloy shown in FIG. 1.

FIG. 3 shows the X-ray image of Fe in the region corresponding to that shown in FIG. 2.

FIG. 4 shows the X-ray image ofNd in the region corresponding to that shown in FIG. 2.

FIG. 5 shows the X-ray image of Dy in the region corresponding to that shown in FIG. 2.

FIG. 6 shows the X-ray image of Ga in the region corresponding to that shown in FIG. 2.

FIG. 7 is an electron image of the R-T-B type alloy shown in FIG. 1.

FIG. 8 shows the X-ray image of Dy in the region corresponding to that shown in FIG. 7.

FIG. 9 shows the X-ray image of Fe in the region corresponding to that shown in FIG. 7.

FIG. 10 shows the X-ray image ofNd in the region corresponding to that shown in FIG. 7.

FIG. 11 is a schematic front view showing the constitution of an apparatus for producing an alloy according to an embodiment of the present invention.

FIG. 12 is a schematic front view showing a casting device provided in the apparatus for producing an alloy.

FIG. 13 is a schematic front view showing a heating device provided in the apparatus for producing an alloy.

FIG. 14 is a schematic side view showing the heating device provided in the apparatus for producing an alloy.

FIG. 15 is a plan schematic view showing opening-closing stages and a storage vessel (container) that are provided in the apparatus for producing an alloy.

FIG. 16 is a schematic front view illustrating the operation of the apparatus for producing an alloy.

FIG. 17 is a schematic front view illustrating the operation of the apparatus for producing an alloy.
FIG. 18 is a schematic front view illustrating the operation of the apparatus for producing an alloy.
FIG. 19 is a schematic front view illustrating the operation of the apparatus for producing an alloy.
FIG. 20 is a schematic side view illustrating the operation of the apparatus for producing an alloy.
FIG. 21 is an electron image of the R-T-B type alloy having no Dy-rich regions.
FIG. 22 shows the X-ray image of Dy in the region corresponding to that shown in FIG. 21.
FIG. 23 shows the X-ray image of Fe in the region corresponding to that shown in FIG. 21.
FIG. 24 shows the X-ray image of Nd in the region corresponding to that shown in FIG. 21.
FIG. 25 is a graph showing the coercive force (Hcj) of the magnets produced in Examples 1 and 2 and Comparative Example 1.

DESCRIPTION OF THE REFERENCE SYMBOLS

[0016] 1: Production apparatus (apparatus for producing alloy); 2: Casting device; 3: Heating device; 4: Storage vessel; 4a: Cooling plate; 5: Container; 6: Chamber; 7: Hopper; 7a: Hopper outlet; 21: Crushing device; 31: Heater; 31c: Opening part; 33: opening-closing stage; 33a: Stage plate; 33b: Opening-closing system; 51: Belt conveyor (movable device); L: Molten alloy; N: flake of cast alloy

BEST MODE FOR CARRYING OUT THE INVENTION

[0017] FIG. 1 is a photograph showing one example of the R-T-B type alloy of the present invention. The photograph is taken when the cross-section of the R-T-B type alloy flake is observed by a scanning electron microscope (SEM). Note that the left hand side is the mold face side in FIG. 1.
The R-T-B type alloy shown in FIG. 1 is produced by an SC method. This R-T-B type alloy has a composition of, in terms of mass ratio, 23% ofNd, 9% of Dy, 1% of B, 1% of Co, and 0.2% of Ga, with the balance being Fe. Note that composition of the R-T-B type alloy (in which R is at least one element selected from Sc, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb, and Lu; T is a transition metal that contains 80% by mass or more of Fe; and B is one that contains 50% by mass or more of boron and also contains at least one element of C and N within a range from 0 to less than 50% by mass) of the present invention is not limited to the abovementioned specific composition and the alloy may have any compositions as long as it is an R-T-B type alloy containing at least Dy.
[0018] The R-T-B type alloy shown in FIG. 1 is composed from an $R_2T_{14}B$ phase (main phase) and an R-rich phase. In FIG. 1, the R-rich phase is indicated in white and the $R_2T_{14}B$ phase (main phase) is indicated in gray. The $R_2T_{14}B$ phase mainly is formed of a columnar crystal and partially formed of an equiaxed crystal. The average crystal grain diameter of the $R_2T_{14}B$ phase in the short axis direction is from 10 to 50 μm. In the $R_2T_{14}B$ phase, a linear R-rich phase extending along the long axis direction of the columnar crystal or a particulated or partially broken R-rich phase is present at the grain boundary and within the grain. The R-rich phase is a non-magnetic phase having a low melting point and enriched with R compared to the overall compositional ratio. The average distance between R-rich phases is from 3 to 10 μm.
[0019] FIGS. 2 to 6 show results of the element distribution analysis (digital mapping) of the R-T-B type alloy shown in FIG. 1 by electron probe microanalysis (EPMA) using a wavelength dispersive X-ray spectrometer (WDS).
FIG. 2 is an electron image of the R-T-B type alloy shown in FIG. 1. The R-rich phase is indicated by a white color and the $R_2T_{14}B$ phase (main phase) is indicated by a gray color.
FIG. 3 is an X-ray image of Fe in the region corresponding to that shown in FIG. 2. It is apparent from FIGS. 2 and 3 that the R-rich phase contains less Fe compared to the main phase.
FIG. 4 is an X-ray image ofNd in the region corresponding to that shown in FIG. 2. It is apparent from FIGS. 2 and 4 that the R-rich phase contains more Nd compared to the main phase.
FIG. 5 is an X-ray image of Dy in the region corresponding to that shown in FIG. 2. It is apparent from FIGS. 2 and 5 that the R-rich phase contains less Dy compared to the main phase.
FIG. 6 is an X-ray image of Ga in the region corresponding to that shown in FIG. 2. It is apparent from FIGS. 2 and 6 that the R-rich phase contains more Ga compared to the main phase.
[0020] FIGS. 7 to 10 show results of the element distribution analysis (digital mapping) using a field emission-electron probe microanalyzer (FE-EPMA).
FIG. 7 is an electron image of the R-T-B type alloy shown in FIG. 1. The R-rich phase is indicated in white and the $R_2T_{14}B$ phase (main phase) is indicated in gray.
FIG. 8 is an X-ray image of Dy in the region corresponding to that shown in FIG. 7. It is apparent from FIGS. 7 and 8 that a Dy-rich region, which is relatively enriched with Dy compared to the R-rich phase and the main phase, is formed close to the R-rich phase. Moreover, it is apparent from FIG. 8 that Dy concentration is lower in the main phase than the Dy-rich region and even lower in the R-rich phase.

FIG. 9 is an X-ray image of Fe in the region corresponding to that shown in FIG. 7. It is apparent from FIGS. 7 and 9 that the R-rich phase contains less Fe compared to the main phase.

FIG. 10 is an X-ray image of Nd in the region corresponding to that shown in FIG. 7. It is apparent from FIGS. 7 and 10 that the R-rich phase contains more Nd compared to the main phase.

(Production method)

[0021] The R-T-B type alloy of the present invention shown in FIG. 1 can be cast by, for example, the SC method that uses the apparatus for producing an alloy shown in FIG. 11.

[Apparatus for producing an alloy]

[0022] FIG. 11 is a schematic front view showing an overall configuration of the apparatus for producing an alloy of the present embodiment.

An apparatus 1 for producing an alloy shown in FIG. 11 (hereinafter, described as "production apparatus 1") is equipped with a casting device 2, a crushing device 21, and a heating device 3 in general. The heating device 3 includes a heater 31 and a container 5. The container 5 includes a storage vessel 4, and an opening-closing stage group 32 provided over the storage vessel 4.

The production apparatus 1 shown in FIG. 11 is equipped with a chamber 6. The chamber 6 includes a casting chamber 6a and a temperature-maintaining storage chamber 6b which is provided below the casting chamber 6a, and which is connected to the casting chamber 6a. The casting device 2 is installed in the casting chamber 6a, and the heating device 3 is installed in the temperature-maintaining storage chamber 6b. In addition, a gate 6e is provided in the temperature-maintaining storage chamber 6b, and the temperature-maintaining storage chamber 6b is closed with the gate 6e except when the container 5 is conveyed outside the temperature-maintaining storage chamber 6b.

Moreover, the casting device 2 is also equipped with the crushing device 21 and a hopper 7 is provided between the casting device 2 and the opening-closing stage group 32. The hopper 7 directs the cast alloy flake onto the opening-closing stage group 32.

[Casting device]

[0023] FIG. 12 is a schematic front view showing the casting device 2 that is provided in the production apparatus 1. The casting device 2 shown in FIG. 12 includes a cooling roll 22 that casts a molten alloy L into a cast alloy M by way of rapidly cooling the molten alloy using a water-cooling system (not shown in the figures); a tundish 23 that supplies the cooling roll 22 with the molten alloy L; and the crushing device 21 that crushes the cast alloy M into a cast alloy flake N. As shown in FIG. 12, the crushing device 21, for example, includes a pair of crushing rolls 21a.

[Heating device]

[0024] FIG. 13 is a schematic front view showing a heating device 3 which is provided in the production apparatus 1, FIG. 14 is a schematic side view thereof, and FIG. 15 is a plan schematic view thereof.

As shown in FIGS. 13 to 15, a heater 31 included in the heating device 3 has a heater cover 31 a, and a main body 31b attached below the heater cover 31a. The heater cover 31a is provided therein in order to release the heat generated from the main body 31b to the direction of the container 5, and in order to prevent the heat from the main body 31b from being released to the casting chamber 6a. Also, if the heater cover 31 a is provided therein, then it can prevent the main body 31b from breaking down in the event of a portion of the molten alloy or the cast alloy unexpectedly falling down thereto.

The heater 31 has an opening part 3 1c, and an outlet 7a of the hopper 7 is disposed in the opening part 31c. Consequently, the flake N of the cast alloy that passes through the hopper 7 and then falls down from the casting device 2 can be supplied to an opening-closing stage group 32 in the container 5 which is provided below the heater 31.

Moreover, the heater 31, as shown in FIGS. 11 and 13, is disposed along the longitudinal direction of a belt conveyor 51 (the moving direction of the container 5), which is provided inside the temperature-maintaining storage chamber 6b. This configuration makes it possible to uniformly maintain the temperature of the flake N of the cast alloy mounted on the opening-closing stage group 32 in the container 5 even when the container 5 moves inside the temperature-maintaining storage chamber 6b.

[0025] The opening-closing stage group 32 included in the heating device 3 is integrated with the storage vessel 4 to form the container 5. That is, the container 5 shown in FIGS. 13 to 15 is formed with the storage vessel 4, and the opening-closing stage group 32 which is provided over the storage vessel 4.

The opening-closing stage group 32 is equipped with a plurality of opening-closing stages 33 that are disposed along the moving direction of the container 5. In addition, guide members 52 are provided around the opening-closing stage group 32, and the guide members 52 prevent the flake N of the cast alloy that drop through the hopper 7 from scattering into the temperature-maintaining storage chamber 6b.

[0026] Each opening-closing stage 33 leaves the flake N of the cast alloy, which are supplied from the casting device 2, mounted thereon to maintain the temperature with the heater 31 at a predetermined period, and drops the flake N of the cast alloy to the storage vessel 4 after the temperature holding time. Each opening-closing stage 33 is equipped with a stage plate 33a, and an opening-closing system 33b which opens or closes the stage plate 33a. Each opening-closing system 33b has a rotating shaft $33b_1$ attached to one side of the stage plate 33a; and a driving unit (not shown in the figures), which rotates the rotating shaft $33b_1$. Each driving unit can freely rotate the rotating shaft $33b_1$ such that the inclination angle of each stage plate 33a can be controlled separately. The inclination angle of each stage plate 33a can be set anywhere in the range of 0° (where the stage plate 33a is horizontal (the position shown in FIG 13 with a two-dot chain line)) to about 90° in the clockwise-direction (where the stage plate 33a is almost vertical (the position shown in FIG 13 with a continuous line)).

[0027] Thus, the opening-closing stage 33 can leave the flake N of the cast alloy mounted on the stage plate 33a during a predetermined temperature holding time by actuating the opening-closing system 33b, and then, can drop the flake N of the cast alloy down into the storage vessel 4 by making the inclination angle of the stage plate 33a larger.
In addition, the opening-closing stage 33 can function as a cover for the storage vessel 4 and this prevents the heat of the heater 31 from reaching the storage vessel 4, thereby preventing the inside of the storage vessel 4 from heating up.
Also, a plurality of cooling plates 4a is provided inside the storage vessel 4.

[0028] Moreover, as shown in FIGS. 13 and 14, the container 5 is mounted on the belt conveyor 51 (movable device). The belt conveyor 51 enables the container 5 to move to the left or right hand side of FIG. 13.

[Alloy casting]

[0029] All FIGS. 16 to 19 are schematic front views illustrating the operation of the apparatus for producing an alloy. As shown in FIG. 16, the container 5 is first moved to where an opening-closing stage 33A (present at the left edge of the opening-closing stage group 32 in the drawing) is located directly under the outlet 7a of the hopper 7. Also, all opening-closing stages 33 are set in a closed state.
Then, flakes N of the cast alloy are prepared by actuating the casting device 2 shown in FIG. 12. A molten alloy L is first prepared in a melting device (not shown in the figures). The temperature of the molten alloy L varies with the types of alloy contents, but it is adjusted within a range of 1,300°C to 1,500°C. The prepared molten alloy L is conveyed to the casting device 2 as it is kept in the refractory crucible 24. Then, the molten alloy L is supplied from the refractory crucible 24 to the tundish 23, and further supplied from the tundish 23 to the cooling roll 22, whereby the molten alloy L is solidified to produce a cast alloy M. After that, the cast alloy M is displaced from the cooling roll 22 to the opposite side of the tundish 23, and is inserted between two rotating crushing rolls 21a, such that the cast alloy M is crushed into flakes N of the cast alloy.

[0030] The average molten alloy supply rate to the cooling roll 22 is 10 g/sec or more, preferably 20 g/sec or more, more preferably 25 g/sec or more, per 1-cm width, and even more preferably 100 g/sec or less per 1-cm width. If the supply rate of molten alloy L is less than 10 g/sec, the molten alloy L may not be thinly wetted and spread on the cooling roll 22 and instead may shrink because of the viscosity of the molten alloy L itself or wettability to the surface of the casting roll 22 and fluctuation of the alloy quality may be brought about. On the other hand, if the average molten alloy supply rate to the cooling roll 22 exceeds 100 g/sec per 1-cm width, cooling on the cooling roll 22 may be insufficient and may cause coarsening of the microstructure, precipitation of α-Fe, or the like.

[0031] The average cooling rate of the molten alloy on the cooling roll 22 is preferably from 100 to 2,000°C/sec. An average cooling rate of 100°C/sec or more will be satisfactory to prevent precipitation of α-Fe or texture coarsening of the R-rich phase or the like. On the other hand, if the average cooling rate is 2,000°C/sec or less, the degree of supercooling will not be excessive and the cast alloy flake can be supplied to the heating device 3 at an appropriate temperature. Moreover, the cast alloy flake does not cool too much, and thus requires no reheating process. Note that the average cooling rate is determined by dividing the difference between the temperature immediately before contact of the molten alloy with the cooling roll and the temperature on detaching from the cooling roll by the time for which the molten alloy is contacted with the cooling roll.

[0032] The average temperature of the cast alloy M on detaching from the cooling roll 22 subtly varies due to a fine difference in the degree of contact between the cast alloy M and the cooling roll 22, fluctuation of the thickness of the cast alloy M, or the like. The average temperature of the cast alloy M on detaching from the cooling roll can be obtained, for example, by scanning the alloy surface in the width direction with a radiation thermometer from the start to finish of casting, thereby measuring the temperature, and averaging the measured values.

[0033] The average temperature of the cast alloy M on detaching from the cooling roll 22 is preferably 100 to 500°C lower, more preferably 100 to 400°C lower, than the solidification temperature of the $R_2T_{14}B$ phase in an equilibrium state of the molten alloy. The melting temperature of the $R_2T_{14}B$ phase is acknowledged to be 1,150°C in the Nd-Fe-B ternary system but varies according to the substitution of Nd by other rare earth elements, the substitution of Fe by other transition elements, and the kind and amount added of any additive elements. If the difference between the average temperature of the cast alloy M on detaching from the cooling roll 22 and the solidification temperature of the $R_2T_{14}B$ phase in an equilibrium state of the cast alloy M is less than 100°C, this may correspond to an insufficient cooling rate. On the other hand, if this difference exceeds 500°C, the supercooling of molten alloy may become excessively large due to too high a cooling rate.

[0034] The average temperature of the cast alloy M on detaching from the cooling roll 22 also varies within the same casting step (tap) and if the variation width is large, this may bring about fluctuation of the microstructure or quality. Therefore, the variation width of temperature within the tap is suitably smaller than 200°C, preferably 100°C or less, more preferably 50°C, even more preferably 20°C.

[0035] The cast alloy flake N preferably has an average thickness of 0.1 to 1 mm. If the average thickness of the flake is less than 0.1 mm, the solidification rate may be excessively increased and the R-rich phase may be dispersed too finely. On the other hand, if the average thickness of the flake exceeds 1 mm, the solidification rate may decrease and this may causes a reduction in the dispersibility of the R-rich phase, precipitation of α-Fe, or the like.

[0036] Next, as shown in FIG. 16, the flakes N of the cast alloy are delivered to the heating device 3 by passing through the hopper 7, and are piled (mounted) on the opening-closing stage 33A which is positioned directly under the outlet 7a of the hopper 7. During this time, the heater 31 is switched on and the flakes N of the cast alloy are kept at a predetermined temperature or heated by the heater 31 immediately after they are piled on the opening-closing stage 33A.

The amount of flakes N of the cast alloy piled on the opening-closing stage 33A may appropriately be adjusted in accordance with the area of the stage plate 33a. However, since the flakes N of the cast alloy are continuously supplied from the casting device 2, they will overflow from the opening-closing stage 33A in time although it also depends on the supply rate. For this reason, the container 5 is moved to the left-hand side in the drawing as shown in FIG. 17 when the piling amount of the flakes N of the cast alloy reaches a predetermined value with respect to the opening-closing stage 33A. Then, another opening-closing stage 33B next to the opening-closing stage 33A on the right side is positioned directly under the outlet 7a of the hopper 7, followed by the flakes N of the cast alloy being piled on the opening-closing stage 33B. After that, in the same way, the container 5 is moved in accordance with the preparation of the flakes N of the cast alloy, and the flakes N of the cast alloy are piled sequentially on the opening-closing stages 33C to 33E.

[0037] The flakes N of the cast alloy piled on each of the opening-closing stages 33A to 33E are kept at a predetermined temperature or heated with the heater 31. It is preferable that the holding temperature be lower than the temperature of the flake N when detaching from the cooling roll (detaching temperature), and specifically, it is preferably within a range of (the detaching temperature - 100°C) to the detaching temperature, and it is more preferably within a range of (the detaching temperature - 50°C) to the detaching temperature. More specifically, the holding temperature is preferably within a range of 600°C to 900°C. When the holding temperature is 600°C or more, the coercive force of an R-T-B type alloy can be sufficiently enhanced. Also, when the holding temperature is 900°C or less, the deposition of α-Fe can be prevented, and the microstructure such as the R-rich phase can be prevented from being coarse.

In addition, when the detaching temperature declines for any reason, the flakes N of the cast alloy can be heated and kept at a predetermined temperature by setting the holding temperature higher than the detaching temperature. It is preferable that the heating range be within 100°C, and more preferably within 50°C. If the heating range is too large, the production efficiency will decline. It should be noted that the coercive force can be improved even when the flakes are kept at 1,000°C. However, such a temperature makes microstructure coarse. Furthermore, the particle distribution or the fluidity of the fine powder when they are finely crushed, and the sintering temperature may unfavorably change. Accordingly, when they are kept at 1,000°C, it is required to consider its influence to subsequent processes.

[0038] Furthermore, the temperature holding time is preferably 30 seconds or more, more preferably 30 seconds to about several hours, and most preferably 30 seconds to about 30 minutes. If the temperature holding time is 30 seconds or more, then the coercive force can be sufficiently enhanced. That is, the flakes of the cast alloy may be subjected to the temperature holding treatment for several hours, but the temperature holding time is preferably 30 minutes or less in terms of the production efficiency.

[0039] Next, as shown in FIG. 18, the container 5 is further moved with respect to the rest of the opening-closing stages 33F to 33J in accordance with the preparation of the flakes N of the cast alloy in the same way, whereby the flakes N of the cast alloy are successively piled on each of the opening-closing stages 33F to 33J. With regard to the flakes N of the cast alloy piled on the opening-closing stages 33A to 33D, they are successively dropped into the storage vessel 4 by sequentially making each opening-closing stage in an opened state as shown in FIG. 18 when the predetermined temperature holding time passes. Once the flakes N of the cast alloy are dropped into the storage vessel 4, the heat of the heater 31 no longer reaches the flakes N of the cast alloy so that the temperature holding treatment is finished.

As described above by referring to FIG. 17, flakes N of the cast alloy are successively mounted on each opening-closing stage, and the different opening-closing stages consequently have time-differences in the starting point to start the temperature holding treatment with respect to the flakes N of the cast alloy on the opening-closing stages. Therefore, it is preferable that the flakes N of the cast alloy be successively dropped into the storage vessel 4 by successively switching each opening-closing stage to an opened state in order to fix the temperature holding time with respect to flakes N of the cast alloy on each opening-closing stage.

The flakes N of the cast alloy that dropped into the storage vessel 4 are in contact with the cooling plate 4a, whereby the heat is absorbed into the cooling plate 4a, and the flakes N of the cast alloy are consequently cooled down.

[0040] FIGS. 19 and 20 show a state in which all opening-closing stages 33 are in an opened state, and the flakes N of the cast alloy are stored in the storage vessel 4. If the casting and crushing processes by the casting device 2 are subsequently conducted after that, the container 5 can be moved to the right hand in the figures while all opening-closing stages 33 are set to a closed state, and the flakes N of the cast alloy are successively mounted on each opening-closing stage 33 in accordance with the preparation of the flakes N of the cast alloy. To the contrary, if the casting and crushing processes by the casting device 2 are terminated, all opening-closing stages 33 are switched to in a closed state to prevent the heat of the heater 31 from reaching the storage vessel 4. Then, the gate 6e of the temperature-maintaining storage chamber 6b is opened, and the container 5 is conveyed outside the chamber 6 to collect the flakes N of the cast alloy, thereby completing the production of flakes N of the cast alloy.

[Cooling rate]

[0041] Next, the cooling rate on producing the R-T-B type alloy will be described.

In the present invention, the cooling rate was controlled so as to achieve the following cooling rates from the solidifying point of the main phase (around 1,170°C), which is a temperature immediately after the solidification, to 600°C, which is lower than the solidifying point of the R-rich phase (around 700°C).

That is, the cooling rate of R-T-B type alloy from 1,000°C to 850°C is set within a range of 100 to 300°C/sec. If the cooling rate from 1,000°C to 850°C is higher than the above range, Dy may not sufficiently diffuse into the main phase. On the other hand, if the cooling rate is lower than the above range, Dy may diffuse excessively, making it impossible to form a Dy-rich region in the main phase.

Also, it is preferable that the cooling rate of R-T-B type alloy from the solidifying point of the main phase to 1,000°C be set within a range of 300 to 2,000°C/sec. By setting the cooling rate from the solidifying point of the main phase to 1,000°C within the above range, an R-T-B type alloy having a Dy-rich region is obtained with high productivity.

In addition, it is preferable that the cooling rate of R-T-B type alloy from 850°C to 600°C be temporarily set to 100°C/sec or less. By temporarily setting the cooling rate from 850°C to 600°C within the above range, Dy contained in the R-rich phase can diffuse into the adjacent main phase sufficiently. Accordingly, an R-T-B type alloy having a Dy-rich region and even higher coercive force can easily be produced.

[0042] The R-T-B type alloy and the flakes of the R-T-B type alloy of the present embodiment are formed close to the R-rich phase and have a Dy-rich region that is relatively enriched with Dy compared to the overall compositional ratio. Accordingly, a rare earth permanent magnet having a high coercive force and excellent magnetic characteristics can be achieved from them.

In other words, the R-T-B type alloy of the present embodiment has a higher coercive force compared to, for example, the R-T-B type alloy shown in FIGS. 21 to 24 having no Dy-rich regions.

[0043] FIGS. 21 to 24 show the results of the element distribution analysis (digital mapping) of the R-T-B type alloy having no Dy-rich region as one example using a field emission-electron probe microanalyzer (FE-EPMA). The R-T-B type alloy shown in FIGS. 21 to 24 is produced by an SC method. This R-T-B type alloy has a composition of, in terms of mass ratio, 23% ofNd, 9% of Dy, 1% of B, 1% of Co, and 0.2% of Ga, with the balance being Fe.

[0044] FIG. 21 is an electron image of the R-T-B type alloy having no Dy-rich regions. The R-rich phase is indicated in white and the $R_2T_{14}B$ phase (main phase) is indicated in gray.

FIG. 22 is an X-ray image of Dy in the region corresponding to that shown in FIG. 21. It is apparent from FIGS. 21 and 22 that this R-T-B type alloy does not have any Dy-rich region that is enriched with Dy compared to the main phase, and that Dy concentration is lower in the R-rich phase than in the main phase.

FIG. 23 is an X-ray image of Fe in the region corresponding to that shown in FIG. 21. It is apparent from FIGS. 21 and 23 that the R-rich phase contains less Fe compared to the main phase.

FIG. 24 is an X-ray image of Nd in the region corresponding to that shown in FIG. 21. It is apparent from FIGS. 21 and 24 that the R-rich phase contains more Nd compared to the main phase.

(Production of R-T-B Type Rare Earth Permanent Magnet)

[0045] For producing the R-T-B type rare earth permanent magnet of the present invention, a fine powder for R-T-B type rare earth permanent magnets is first produced from the R-T-B type alloy of the present invention. The fine powder for R-T-B type rare earth permanent magnets of the present invention is obtained, for example, by a method of performing hydrogen cracking of a flake formed of the R-T-B type alloy of the present invention by hydrogen absorption and then pulverizing the flake by using a grinder such as a jet mill. In the hydrogen cracking here, for example, a hydrogen absorption step of keeping the flake in a hydrogen atmosphere under a predetermined pressure is preferably performed in advance.

Then, the obtained fine powder for R-T-B type rare earth permanent magnets is, for example, press-shaped by a shaping machine or the like in a transverse magnetic field and sintered in vacuum, whereby an R-T-B type rare earth permanent magnet is obtained.

[0046] The fine powder for an R-T-B type rare earth permanent magnet and the R-T-B type rare earth permanent magnet of the present embodiment are produced from the R-T-B type alloy of the present invention. Accordingly, they will have a high coercive force and excellent magnetic characteristics.

[Example 1]

[0047] Starting metals formulated to have an alloy composition of, in terms of mass ratio, 23% ofNd, 9% of Dy, 0.98% of B, 1% of Co, and 0.2% of Ga, with the balance being Fe were weighed and then melted in an alumina crucible in an argon gas atmosphere at 1 atm by using a high-frequency melting furnace to produce a molten alloy. Then this molten alloy was supplied to the casting device in the production apparatus shown in FIG. 11 and was cast by the SC method. The rotating speed of the cooling roll at the casting was 1.3 m/s, the average molten alloy supply rate to the cooling roll was 30 g/sec per 1-cm width, and the average temperature of the cast alloy ingot on detaching from the cooling roll was 850°C.

The cooling rate of this alloy was 700°C/sec from the solidifying point of the main phase to 1,000°C, 200°C/sec from 1,000°C to 850°C, and 50°C/sec from 850°C to 780°C. Thereafter, the alloy was kept at a temperature around 780°C for 300 seconds on an opening-closing stage using the production apparatus shown in FIG. 11, and then cooled down to 600°C or less at a cooling rate of 0.1°C/sec to produce flakes of the R-T-B type alloy of Example 1. The average thickness of the alloy at this point was 0.3 mm.

[Example 2]

[0048] A molten alloy was produced by using the same starting metals and the same apparatus as in Example 1. Then the obtained molten alloy was cast using the same casting device as in Example 1. The rotating speed of the cooling roll at the casting was 0.87 m/s, the average molten alloy supply rate to the cooling roll was 30 g/sec per 1-cm width, and the average temperature of the cast alloy ingot on detaching from the cooling roll was 880°C.

The cooling rate of this alloy was 700°C/sec from the solidifying point of the main phase to 1,000°C, 200°C/sec from 1,000°C to 850°C, and 10°C/sec from 850°C to 780°C. Thereafter, the alloy was cooled down to 600°C or less at a cooling rate of 0.1°C/sec using the production apparatus shown in FIG. 11 without employing an opening-closing stage to produce flakes of the R-T-B type alloy of Example 2. The average thickness of the alloy at this point was 0.45 mm.

[0049] The flakes of the R-T-B type alloys obtained in Examples 1 and 2 were subjected to element distribution analysis (digital mapping) (surface analysis) using an electron probe microanalyzer equipped with a wavelength dispersive X-ray spectrometer (WDS-EPMA), and a field emission-electron probe microanalyzer (FE-EPMA). As a result, both flakes of the R-T-B type alloy obtained in Examples 1 and 2 were found to form a Dy-rich region, which was enriched with Dy compared to the R-rich phase and the main phase, near the R-rich phase. Moreover, in both the flakes of the R-T-B type alloy obtained in Examples 1 and 2, the Dy concentration was lower in the main phase than in the Dy-rich region and even lower in the R-rich phase.

(Comparative Example 1)

[0050] A molten alloy was produced by using the same starting metals and the same apparatus as in Example 1. Then the obtained molten alloy was cast using the same casting device as in Example 1 to produce flakes of the R-T-B type alloy of Comparative Example 1. The rotating speed of the cooling roll at the casting was 0.65 m/s, the average molten alloy supply rate to the cooling roll was 15 g/sec per 1-cm width, and the average temperature of the cast alloy ingot on detaching from the cooling roll was 700°C.

The cooling rate of this alloy was 700°C/sec from the solidifying point of the main phase to 1,000°C, 400°C/sec from 1,000°C to 700°C, and 10°C/sec from 700°C to 600°C. Thereafter, the alloy was cooled down to 600°C or less at a cooling rate of 0.1°C/sec using the production apparatus shown in FIG. 11 without employing an opening-closing stage. The average thickness of the alloy at this point was 0.30 mm.

[0051] The flakes of the R-T-B type alloy obtained in Comparative Example 1 were subjected to element distribution analysis (digital mapping) (surface analysis) using WDS-EPMA and the FE-EPMA. As a result, the flakes of the R-T-B type alloy obtained in Comparative Example 1 were found not to form any Dy-rich regions that were enriched with Dy compared to the overall compositional ratio. One possible cause for this result may be that, in Comparative Example 1, the temperature of the cast alloy ingot on detaching from the cooling roll was low and the alloy cooled down too rapidly on the cooling roll, making the cooling rate of the alloy from 1000°C to 700°C too high. Accordingly, Dy and Nd possibly did not diffuse sufficiently and concentration gradients thereof were not formed.

[0052] Next, magnets were produced as follows using the flakes of R-T-B type alloys obtained in Examples 1 and 2 and Comparative Example 1.

The flakes of R-T-B type alloys obtained in Examples 1 and 2 and Comparative Example 1 were first subjected to hydrogen cracking. The hydrogen cracking was carried out by the following method. The flakes of the R-T-B type alloys were made to absorb hydrogen in a hydrogen atmosphere with a pressure of 2 atm, and then were heated to 500°C in vacuum to dehydrogenate. Thereafter, 0.07% by mass of a zinc stearate was added thereto and the resultant was pulverized by a jet mill using a nitrogen gas stream. The powder obtained by the pulverization had an average grain size of about 5.0 μm as measured by laser diffraction.

[0053] Next, the obtained powder material was press-shaped by a shaping machine in a transverse magnetic field at a shaping pressure of 0.8 t/cm^2 in a 100% nitrogen atmosphere to obtain a powder compact. The obtained powder compact was heated from room temperature in a vacuum of 1.33×10^{-5} hPa and was held at 500°C for 1 hour and then at 800°C for 1 hour to remove zinc stearate and the remaining hydrogen. Then the resulting powder compact was heated to a sintering temperature of 1,030°C and was held there for 3 hours to produce a sintered body. Thereafter, the obtained sintered body was further heat-treated for 1 hour at 800°C and then 530°C in an argon atmosphere. As a result, 10 magnets were obtained in both Examples 1 and 2, and 5 magnets were obtained in Comparative Example 1.

The magnetic characteristics of the magnets obtained in Examples 1 and 2 and Comparative Example 1 were measured by a direct current BH curve tracer. The results are shown in Table 1 and FIG. 25. FIG. 25 is a graph showing the coercive force (Hcj) of the magnets produced in Examples 1 and 2 and Comparative Example 1. In the graph, the vertical axis indicates the coercive force of the respective Examples, which are shown in the horizontal axis. In FIG. 25, the symbol o indicates the coercive force of the magnets produced in Examples 1 and 2 and the symbol A indicates the coercive force of the magnets produced in Comparative Example 1.

[0054]

[Table 1]

	Hcj (kOe)	Hk/Hcj (%)	BH_{max} (MGOe)	Br (kG)
Example 1	33.34	56.40%	32.76	11.54
	34.19	54.87%	32.61	11.54
	33.69	56.39%	33.20	11.62
	33.99	56.73%	33.36	11.63
	34.10	56.43%	33.27	11.63
	33.35	55.58%	32.41	11.47
	33.18	57.28%	33.27	11.61
	33.44	57.95%	33.72	11.71
	33.67	56.62%	33.08	11.58
	33.13	57.53%	33.22	11.61
Example 2	32.61	58.71%	33.26	11.64
	33.07	58.03%	32.96	11.58
	33.31	58.49%	33.66	11.68
	33.74	57.79%	33.15	11.61
	33.01	58.33%	33.03	11.58
	33.31	58.09%	32.82	11.60
	33.31	58.84%	32.87	11.57
	33.69	57.53%	33.00	11.60
	33.47	59.14%	33.35	11.65
	33.04	60.06%	33.22	11.66

(continued)

	Hcj (kOe)	Hk/Hcj (%)	BH_{max} (MGOe)	Br (kG)
Comparative Example 1	31.44	52.30%	32.22	11.45
	31.73	52.22%	32.73	11.54
	31.28	53.54%	32.73	11.53
	31.38	53.08%	33.50	11.70
	31.42	52.93%	32.92	11.57

[0055] In Table 1, "$(BH)_{max}$" indicates the maximum magnetic energy product, "Br" indicates the residual magnetic flux density, "Hcj" indicates the coercive force, and "Hk/Hcj" indicates the hysteresis squareness.

[0056] As shown in Table 1 and FIG. 25, the magnets obtained in Examples 1 and 2 had higher coercive forces "Hcj" compared to those of the magnets obtained in Comparative Example 1 that are produced from the R-T-B type alloy with no Dy-rich regions being formed. Such differences in the coercive force of the magnets were caused by the distribution of elemental concentration therein which originated when they were in the alloy state and which continued to have an influence even after the alloys are crushed and sintered to produce the magnets. One possible reason for this is that since the Dy-rich regions are present in the alloys of the present invention and they also remain in the crystal grains of magnets, there is only a small amount of Dy remains in the R-rich phase, which does not contribute efficiently for the improvement of coercive force.

Claims

1. An R-T-B type alloy (wherein R is at least one element selected from Sc, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb, and Lu; T is a transition metal that contains 80% by mass or more of Fe; and B is one that contains 50% by mass or more of boron (B) and also contains at least one element of C and N within a range from 0 to less than 50% by mass) which is a raw material for use in a rare earth-based permanent magnet and contains at least Dy, said R-T-B type alloy comprising:

   a main phase such as an $R_2T_{14}B$ phase exhibiting magnetic properties;
   an R-rich phase that is enriched with R compared to the overall alloy compositional ratio; and
   a Dy-rich region that is formed close to the R-rich phase and enriched with Dy compared to the compositional ratio.

2. The R-T-B type alloy according to Claim 1, wherein a Dy concentration is lower in the main phase than in the Dy-rich region and is lower in the R-rich phase than in the main phase.

3. The R-T-B type alloy according to Claim 1 or 2, wherein the alloy is a flake having an average thickness from 0.1 to 1 mm produced by a strip casting method.

4. A method for producing an R-T-B type alloy according to any one of Claims 1 to 3, said method comprising the steps of:

   producing a flake having an average thickness from 0.1 to 1 mm; and
   supplying molten alloy to a cooling roll at an average rate of 10 g/sec or more per 1-cm width.

5. The method for producing an R-T-B type alloy according to Claim 4, wherein an R-T-B type alloy flake rolled out of the cooling roll is maintained at a temperature from 600 to 900°C for 30 seconds or more.

6. A fine powder for an R-T-B type rare earth permanent magnet that is produced from the R-T-B type alloy according to any one of Claims 1 to 3 or from the R-T-B type alloy produced by the method for producing an R-T-B type alloy according to Claim 4 or 5.

7. An R-T-B type rare earth permanent magnet produced from the fine powder for an R-T-B type rare earth permanent magnet according to Claim 6.

FIG. 1

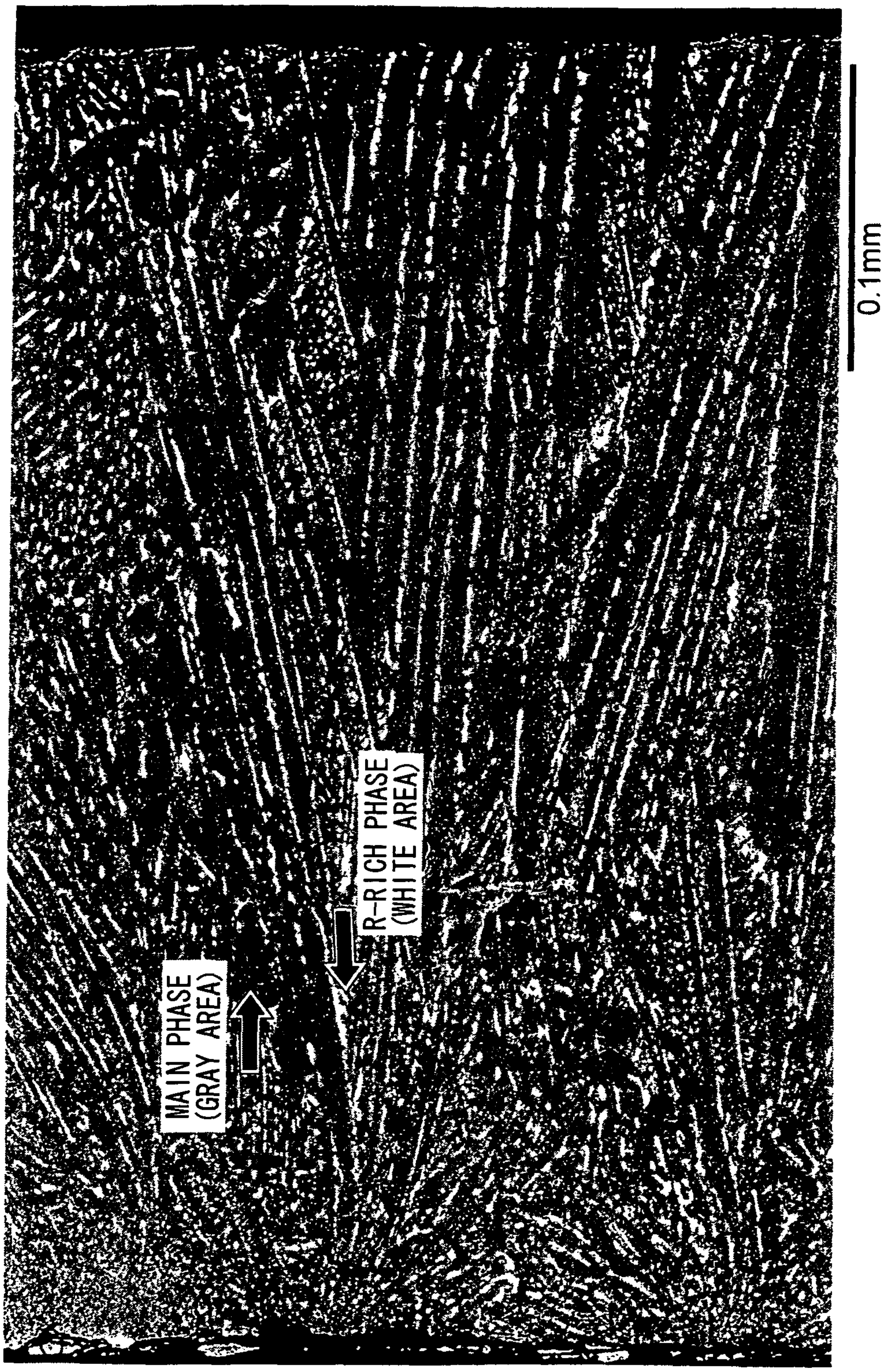


FIG. 2

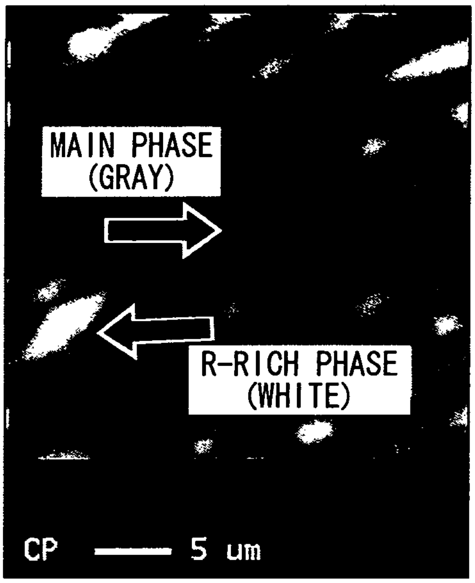

MAIN PHASE
(GRAY)
R-RICH PHASE
(WHITE)
CP
5 um

FIG. 3

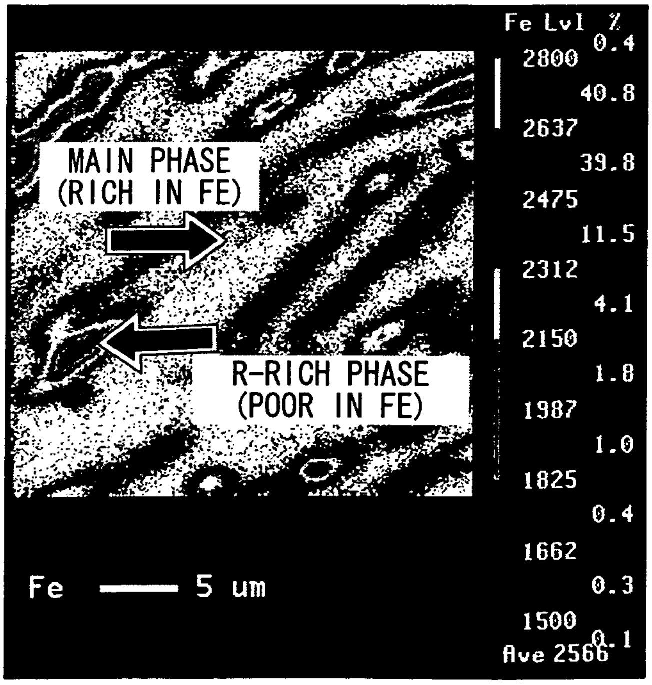

Fe Lvl %
2800
0.4
40.8
2637
39.8
2475
11.5
2312
4.1
2150
1.8
1987
1.0
1825
0.4
1662
0.3
1500
0.1
Ave 2566
MAIN PHASE
(RICH IN FE)
R-RICH PHASE
(POOR IN FE)
Fe
5 um

FIG. 4

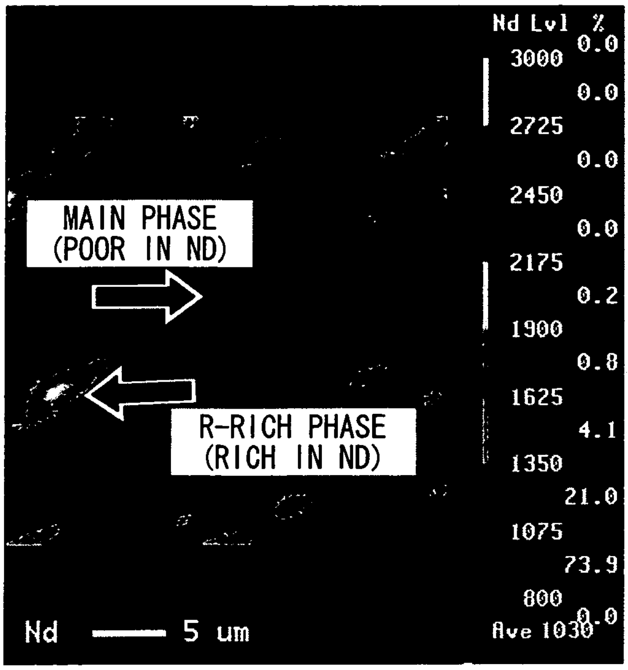

Nd Lvl %
3000
2725
2450
2175
1900
1625
1350
1075
800
Ave 1030
0.0
0.0
0.0
0.0
0.2
0.8
4.1
21.0
73.9
0.0
MAIN PHASE (POOR IN ND)
R-RICH PHASE (RICH IN ND)
Nd
5 um

FIG. 5

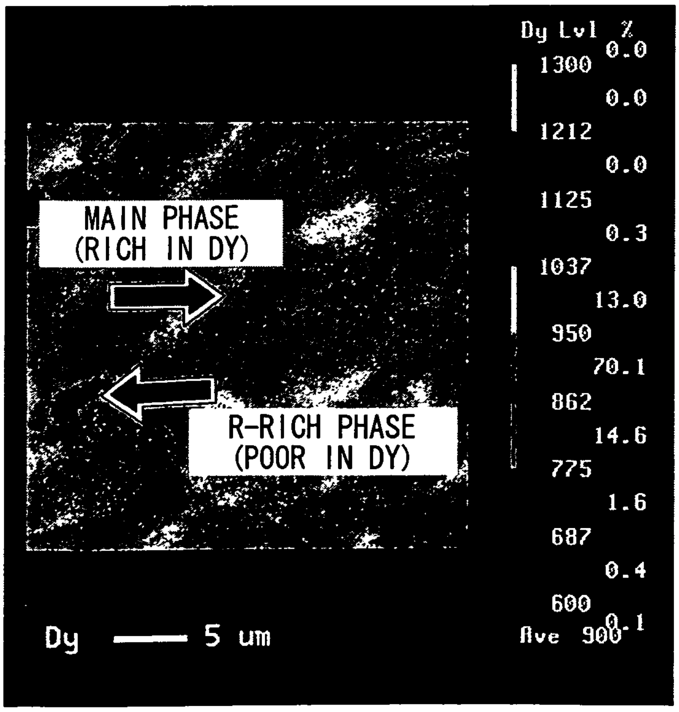

Dy Lvl %
1300
1212
1125
1037
950
862
775
687
600
Ave 900
0.0
0.0
0.0
0.3
13.0
70.1
14.6
1.6
0.4
0.1
MAIN PHASE (RICH IN DY)
R-RICH PHASE (POOR IN DY)
Dy
5 um

FIG. 6

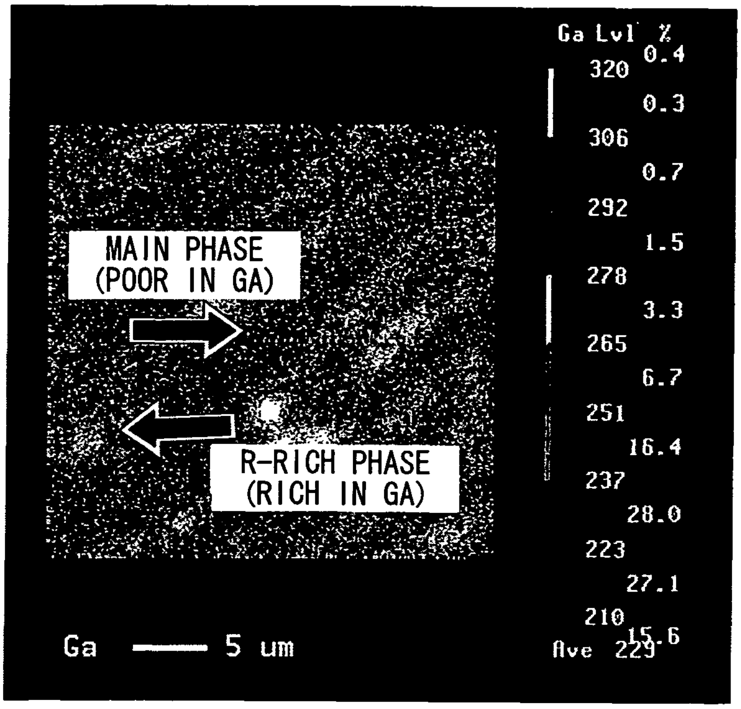

Ga Lvl %
320
0.4
0.3
306
0.7
292
1.5
278
3.3
265
6.7
251
16.4
237
28.0
223
27.1
210
15.6
Ave 229
MAIN PHASE (POOR IN GA)
R-RICH PHASE (RICH IN GA)
Ga
5 um

FIG. 7

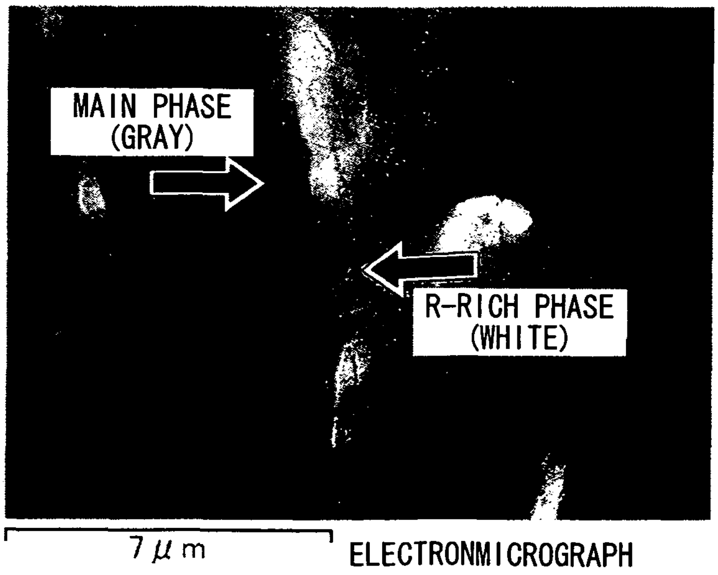

MAIN PHASE (GRAY)
R-RICH PHASE (WHITE)
7 μm
ELECTRONMICROGRAPH

FIG. 8

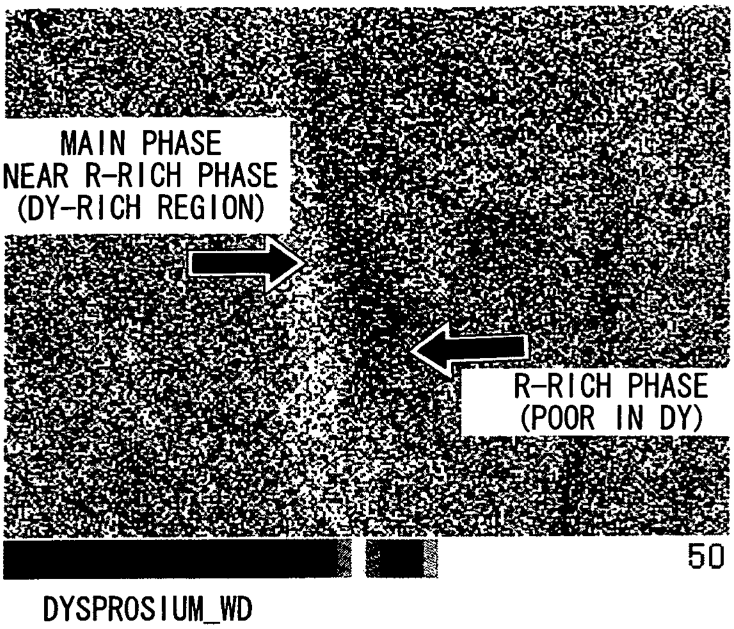

MAIN PHASE
NEAR R-RICH PHASE
(DY-RICH REGION)
R-RICH PHASE
(POOR IN DY)
50
DYSPROSIUM_WD

FIG. 9

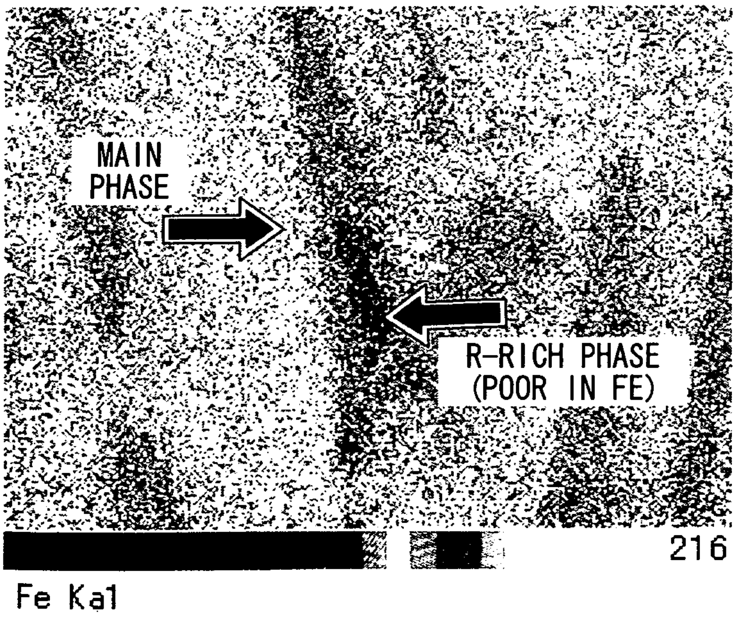

MAIN
PHASE
R-RICH PHASE
(POOR IN FE)
216
Fe Ka1

FIG. 10

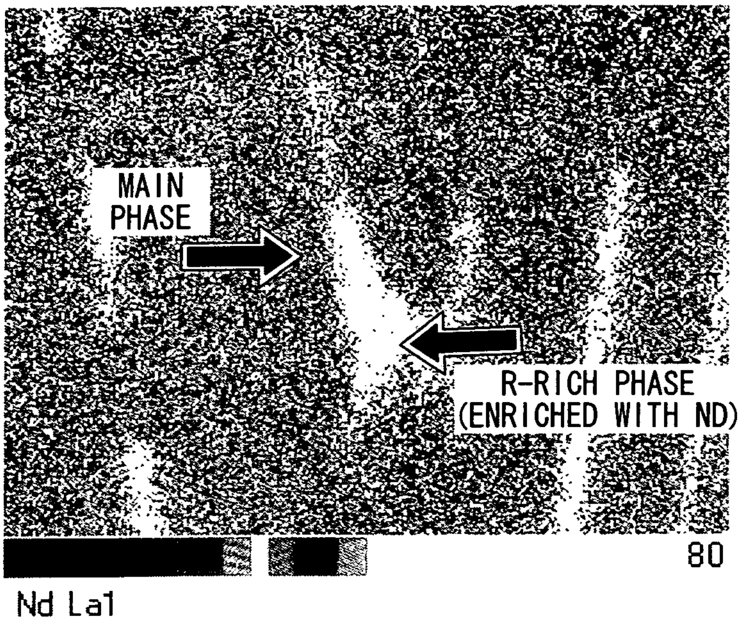


FIG. 11

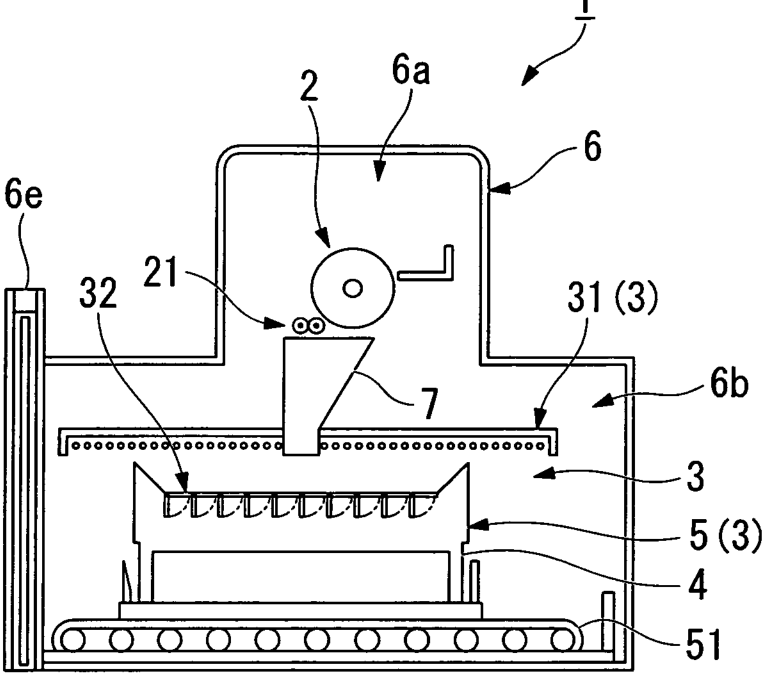


FIG. 12

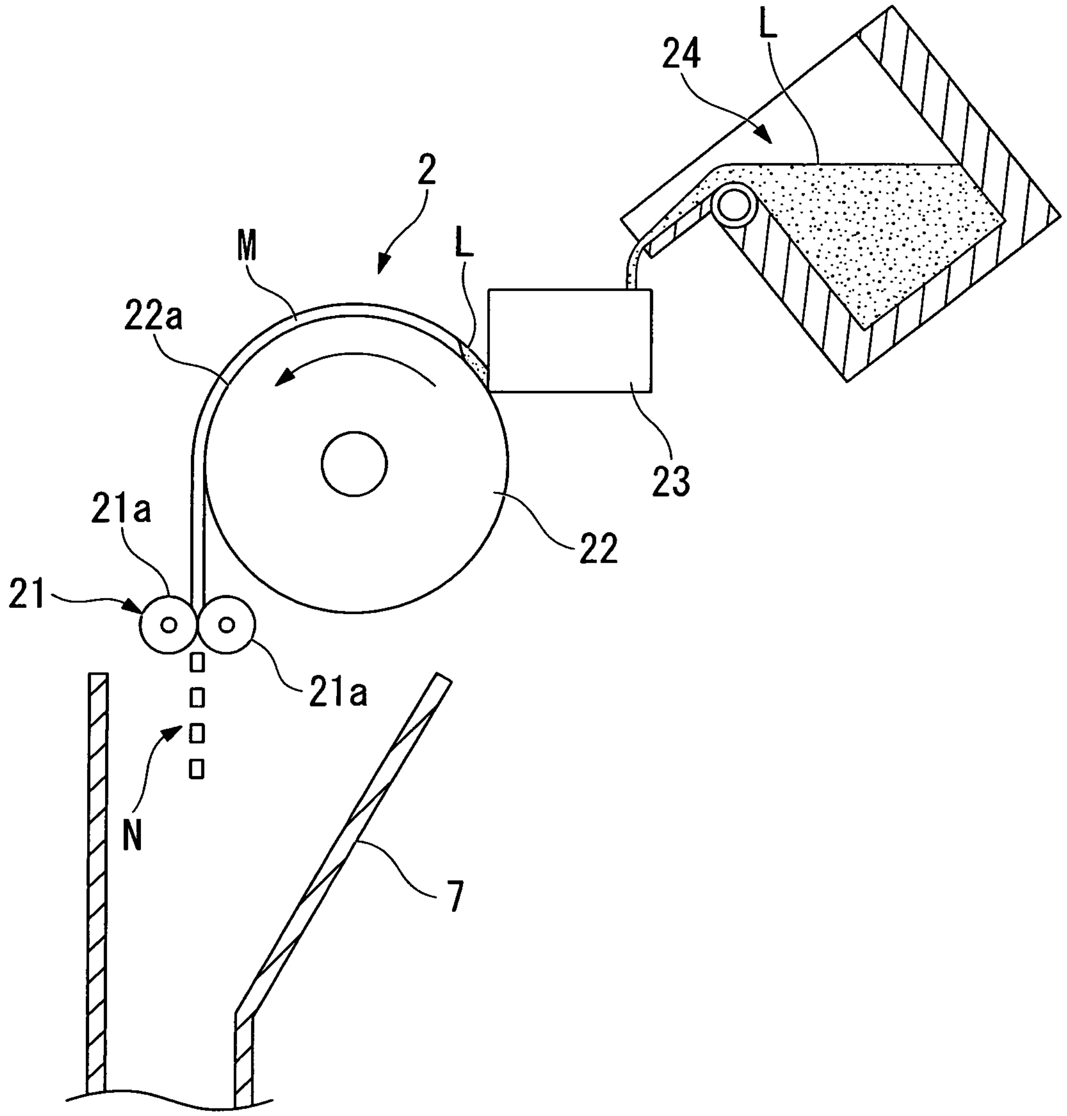


FIG. 13

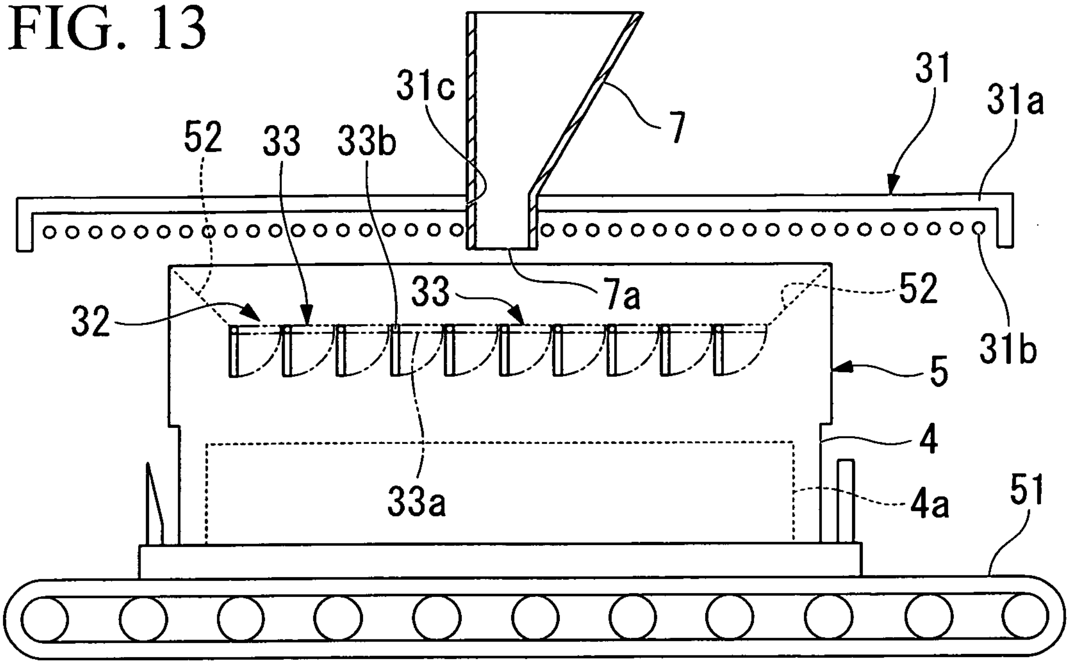

31c
7
31
31a
52
33
33b
33
7a
52
32
31b
5
4
33a
4a
51

FIG. 14

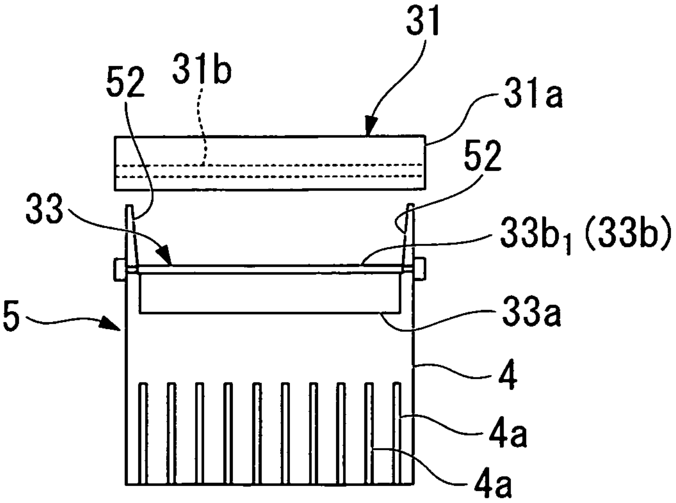

31
52
31b
31a
52
33
33b1 (33b)
5
33a
4
4a
4a

FIG. 15

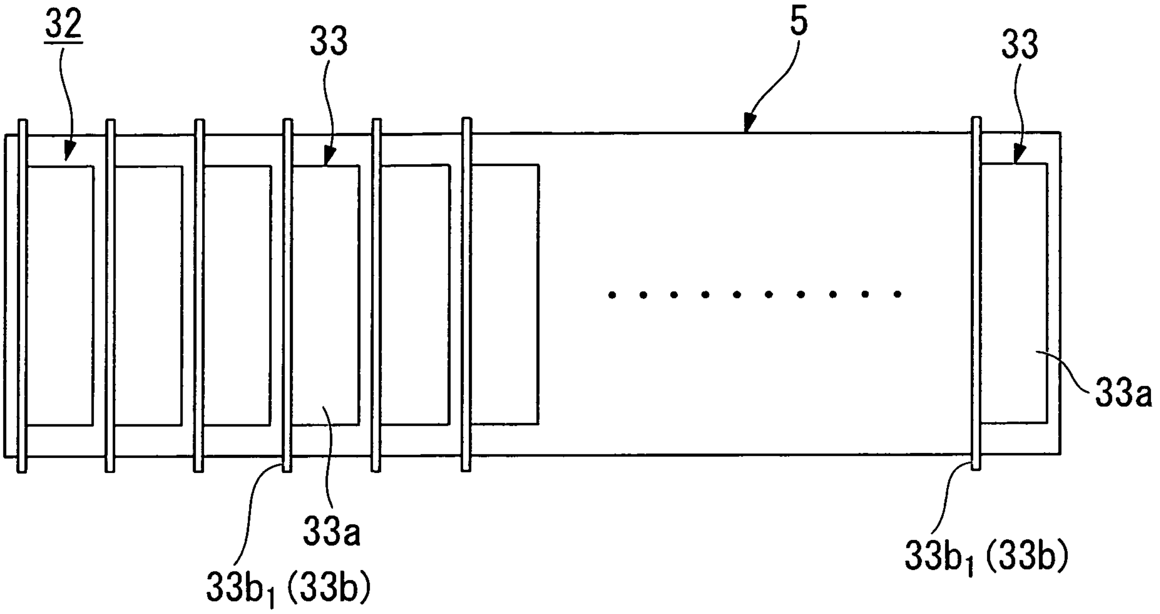


FIG. 16

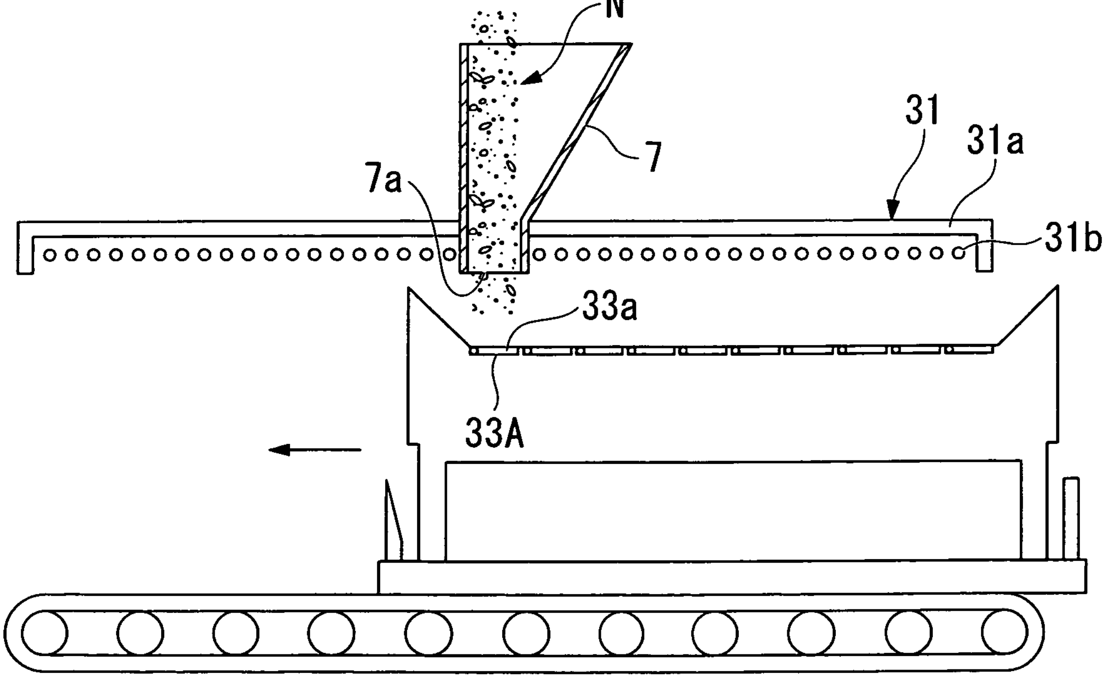


FIG. 17

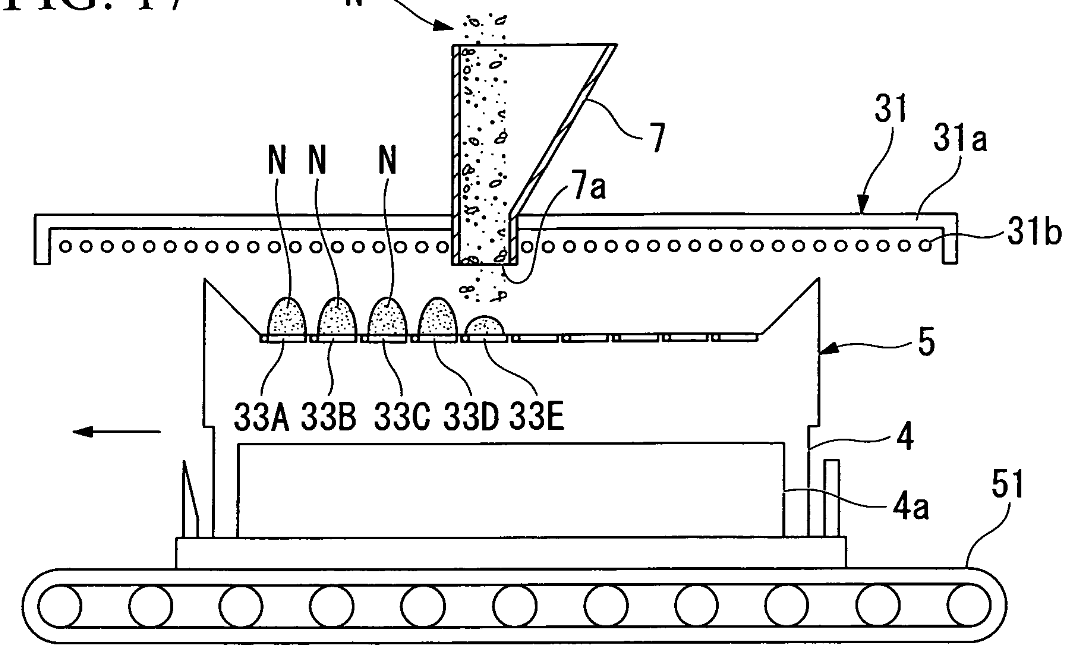

N
7
31
31a
7a
N N N
31b
5
33A 33B 33C 33D 33E
4
4a
51

FIG. 18

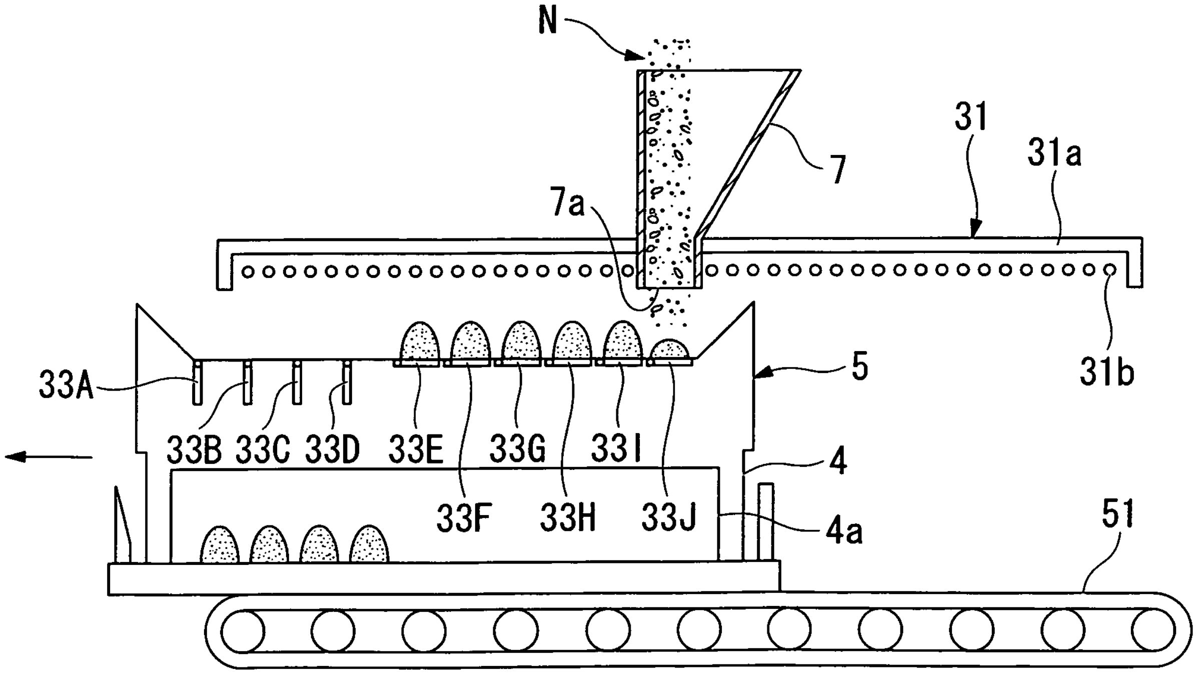

N
31
7
31a
7a
5
31b
33A
33B 33C 33D 33E 33G 33I
4
33F 33H 33J
4a
51

FIG. 19

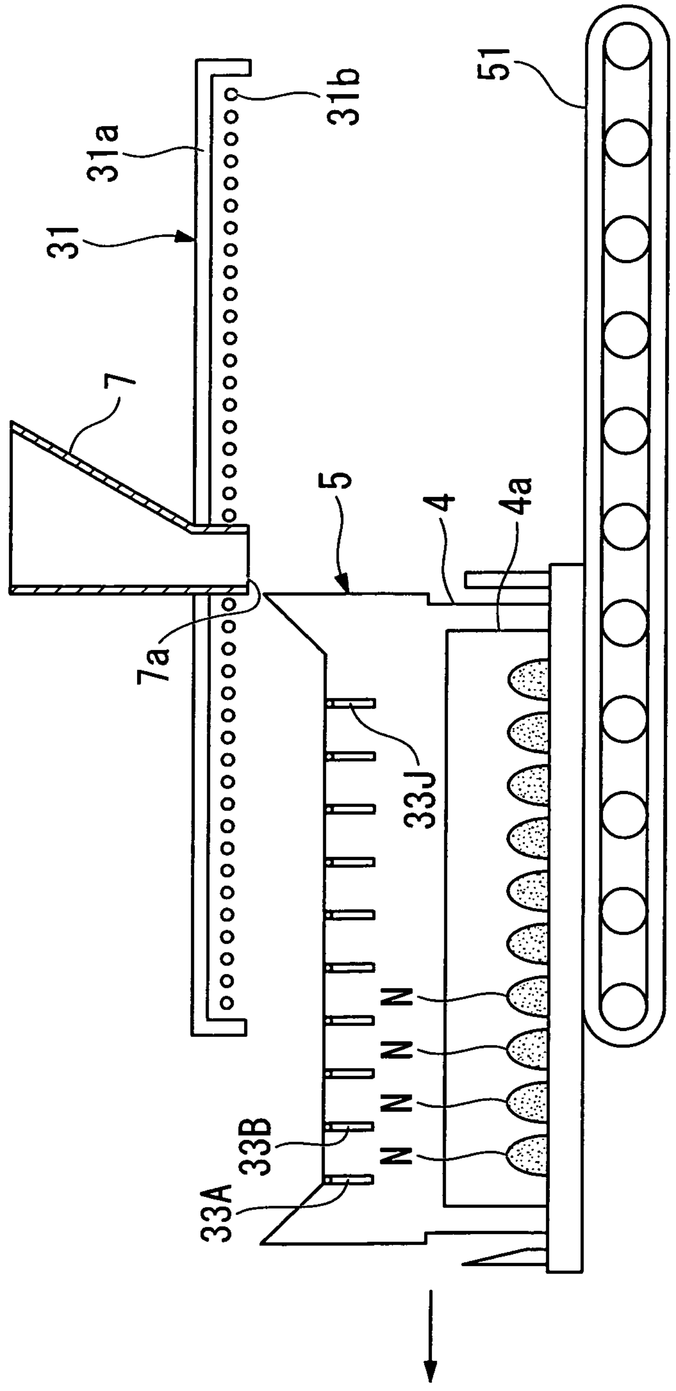


FIG. 20

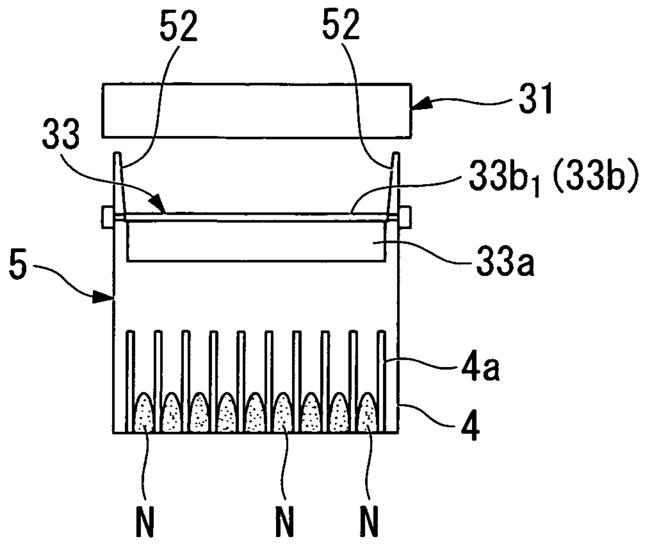


FIG. 21

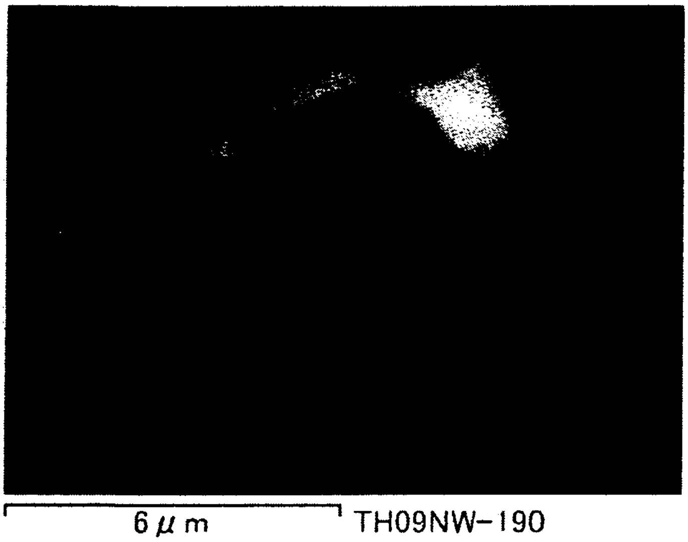


FIG. 22

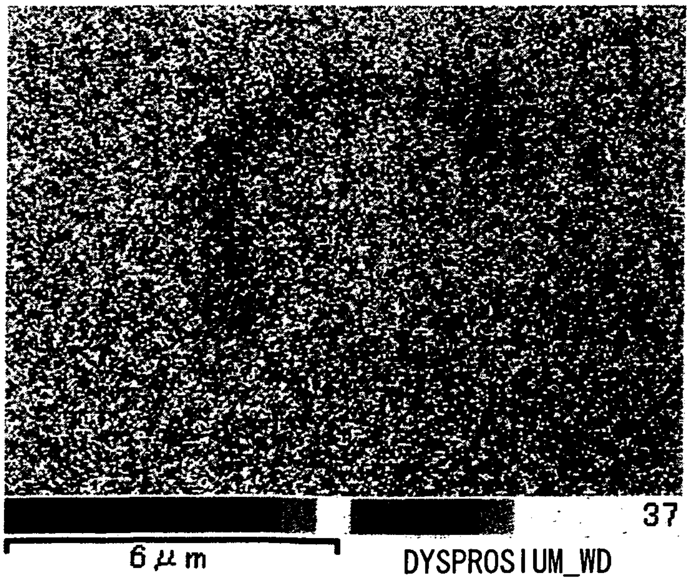


FIG. 23

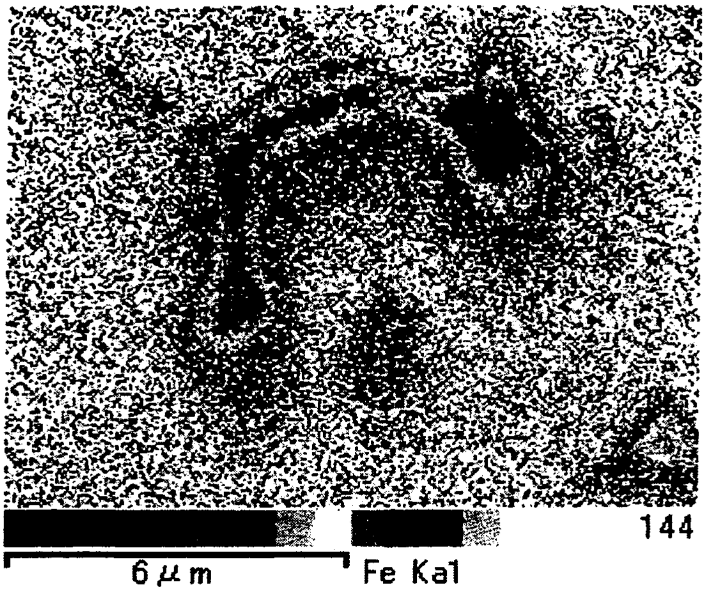


FIG. 24

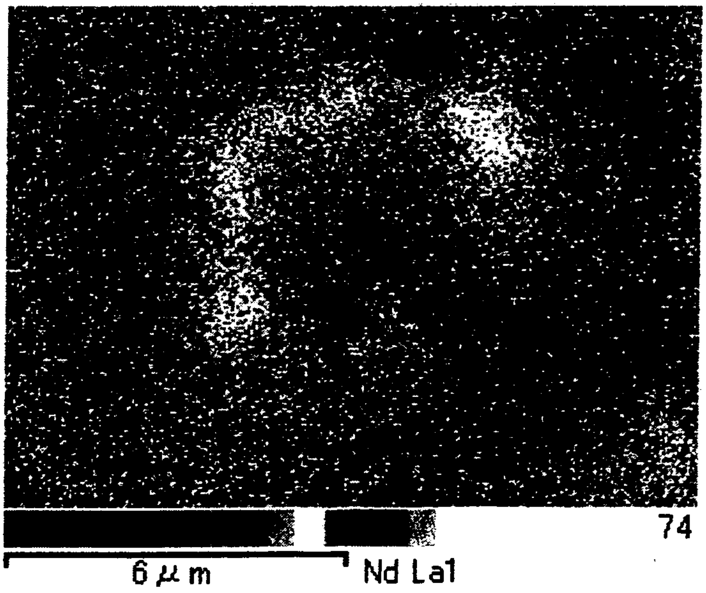
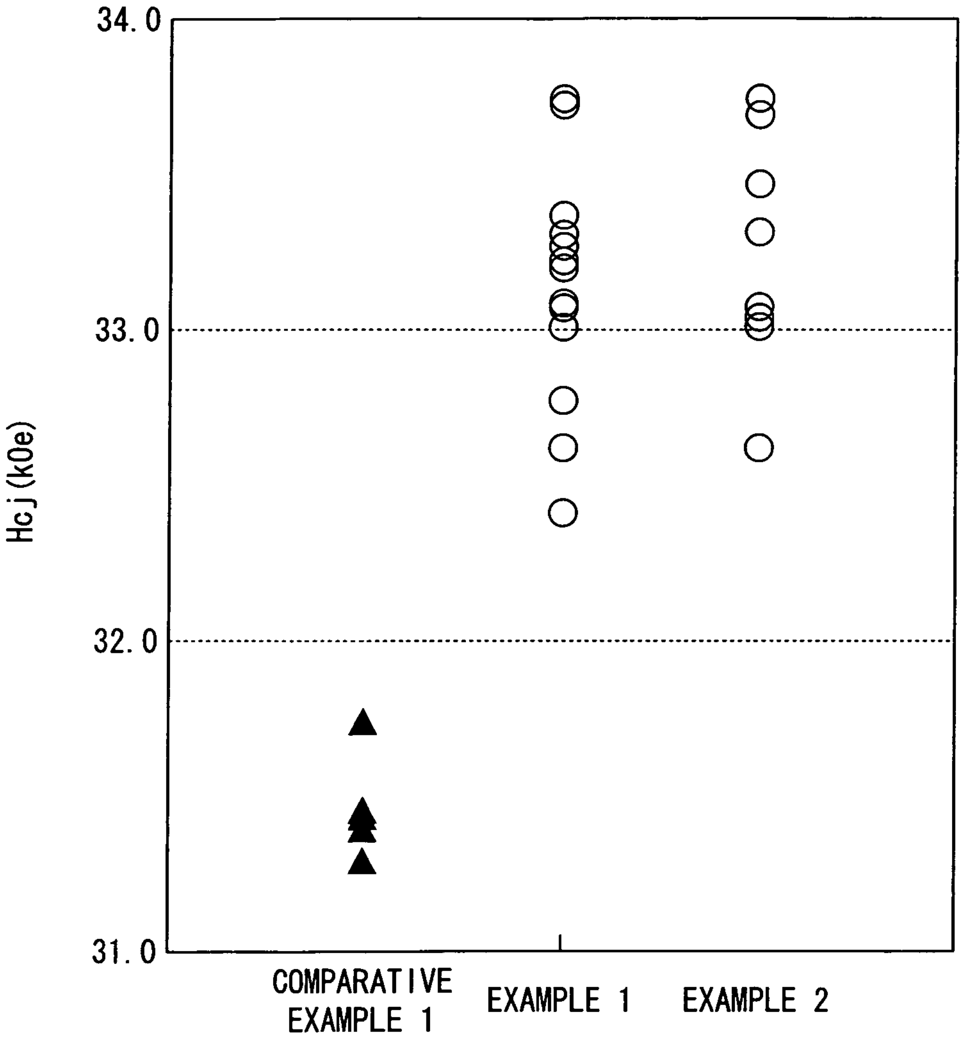


FIG. 25


34.0
33.0
32.0
31.0
Hcj (kOe)
COMPARATIVE EXAMPLE 1
EXAMPLE 1
EXAMPLE 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2008/051186

A. CLASSIFICATION OF SUBJECT MATTER
C22C38/00(2006.01)i, *B22D11/06*(2006.01)i, *B22F1/00*(2006.01)i, *B22F9/04*(2006.01)i, *C21D6/00*(2006.01)i, *C22C33/02*(2006.01)i, *H01F1/053*(2006.01)i, *H01F1/08*(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C22C38/00, B22D11/06, B22F1/00, B22F9/04, C21D6/00, C22C33/02, H01F1/053, H01F1/08

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2008
Kokai Jitsuyo Shinan Koho	1971-2008	Toroku Jitsuyo Shinan Koho	1994-2008

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 03/001541 A1 (Sumitomo Special Metals Co., Ltd.), 03 January, 2003 (03.01.03), (Examples) & US 2004/163737 A1 & DE 10296960 T5	1-7
X	JP 11-315357 A (Showa Denko Kabushiki Kaisha), 16 November, 1999 (16.11.99), (Example 6) (Family: none)	1-7
X A	JP 2003-226944 A (Sumitomo Special Metals Co., Ltd.), 15 August, 2003 (15.08.03), Par. No. [0008]; examples & US 2006/16515 A1 & EP 1479787 A1 & WO 03/66922 A1	1,2,6,7 3-5

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

*	Special categories of cited documents:
"A"	document defining the general state of the art which is not considered to be of particular relevance
"E"	earlier application or patent but published on or after the international filing date
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O"	document referring to an oral disclosure, use, exhibition or other means
"P"	document published prior to the international filing date but later than the priority date claimed
"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&"	document member of the same patent family

Date of the actual completion of the international search
19 March, 2008 (19.03.08)

Date of mailing of the international search report
01 April, 2008 (01.04.08)

Name and mailing address of the ISA/
Japanese Patent Office

Facsimile No.

Authorized officer

Telephone No.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2008/051186

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2006-310660 A (Neomax Co., Ltd.), 09 November, 2006 (09.11.06), Par. No. [0008] (Family: none)	1-7
P,A	JP 2007-119882 A (Showa Denko Kabushiki Kaisha), 17 May, 2007 (17.05.07), (Example 1) & US 2007/95438 A1 & EP 1780736 A1	1-7

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- JP HEI521219 B **[0008]**
- WO 2003001541 A **[0008]**
- WO 2005031023 A **[0008]**
- JP 2003188006 A **[0008]**
- JP 2004043291 A **[0008]**

Non-patent literature cited in the description

- **HIROYUKI TOMIZAWA.** *Journal of the Japan Society of Powder and Powder Metallurgy,* March 2005, vol. 52 (3), 158-163 **[0008]**