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(54) **Rare-earth magnet powder material**

Anisotropes R-T-B-M Magnetpulver

Matériau en poudre pour aimant à base de terre-rare

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(73) Proprietor: **MITSUBISHI MATERIALS
CORPORATION**
Chiyoda-ku, Tokyo (JP)

(72) Inventors:
• **Nakayama, Ryoji, c/o Chuo-kenkyusho**
Omiya-shi, Saitama-ken (JP)

• **Takeshita, Takuo, c/o Chuo-kenkyusho**
Omiya-shi, Saitama-ken (JP)
• **Ishii, Yoshinari, c/o Chuo-kenkyusho**
Omiya-shi, Saitama-ken (JP)

(74) Representative:
Baverstock, Michael George Douglas et al
BOULT WADE TENNANT,
27 Farnival Street
London EC4A 1PQ (GB)

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Description

The present invention relates to a rare-earth magnet powder material excellent in an isotropy, which comprises any of the rare-earth elements including Y (hereinafter referred to as "R"), Fe or a component in which part of the Fe is substituted by Co (hereinafter referred to as "T") and B as the main components, and further containing one or more of Si, Ga, Zr, Nb, Mo, Hf, Ta, W, Al, Ti and V (hereinafter referred to as "M") in an amount of from 0.001 to 5.0 atomic %, of which the main phase is an $R_2T_{14}B$ -type intermetallic compound phase.

An R-Fe-B-M magnet material powder available by homogenizing an R-T-B-M raw material alloy, of which the main phase is an $R_2T_{14}B$ -type intermetallic compound phase, with R, T and B as the main components, further containing M in an amount of from 0.001 to 5.0 atomic % by holding same in an Ar atmosphere at a temperature of from 600 to 1,200°C, or heating the R-T-B-M raw material alloy, without homogenizing, in H_2 gas or a mixed H_2 /inert gas atmosphere from the room temperature, holding same at a temperature of from 500 to 1,000°C to cause occlusion of H_2 , then dehydrogenating same by holding same at a temperature of from 500 to 1,000°C in a vacuum atmosphere or an inert gas atmosphere, then cooling and crushing same is known to have an excellent anisotropy and to have a structure comprising the recrystallized fine aggregate structure of the $R_2T_{14}B$ -type intermetallic compound phase of an average recrystallization grain size of from 0.05 to 20 μm (See Japanese Patent Provisional Publication No. 3-129,702, Japanese Patent Provisional Publication No. 3-129,703, Japanese Patent Provisional Publication No. 4-133,406, and Japanese Patent provisional publication No. 4-133,407).

EP-A-0 304 054 describes the preparation of rare earth-iron-boron magnet powders having an average particle size of 2.0 to 500 μm and which powders contain a recrystallized grain structure containing a $R_2Fe_{14}B$ intermetallic compound phase where R represents a rare-earth element, the average crystal grain size of the recrystallized grains being of 0.05 μm to 50 μm . The powder may also further comprise elements M as described herein.

There have been reported some Dulk sinter magnets produced from R-Fe-B-M magnet powder materials having a maximum energy product exceeding 50 MGOe. Although, from the original magnetic properties of the material, a maximum energy product of about 25 MGOe is expected even in a bonded magnet available by using an R-Fe-B-M magnet powder material, bonded magnets and hot-pressed magnets (hereinafter referred to as a "full-density magnet") actually manufactured with the use of the above-mentioned anisotropic R-Fe-B-M magnet powder obtained through H_2 occlusion and dehydrogenation have shown insufficient properties. anisotropy than conventional ones with the use of this R-Fe-B-M magnet powder, and the following discoveries were made.

The grain size of anisotropic R-Fe-B-M magnet powders having the recrystallized fine aggregate structure of the $R_2T_{14}B$ -type intermetallic compound phase of an average recrystallization grain size of from 0.05 to 20 μm obtained through conventional H_2 occlusion and dehydrogenation exerts an important effect on the magnetic properties of bonded magnets and full-density magnets made from them. Bonded magnets and full-density magnets prepared by the use of an anisotropic R-Fe-B-M magnet powder having an average grain size within a range of from 5 to 200 μm , magnetic anisotropy is remarkably improved, and furthermore, during the process of forming in a magnetic field for imparting anisotropy, the an isotropic magnet powder is given a sufficient orientation in an oriented magnetic field within 1.5 times iH_c of the anisotropic magnet powder.

The present invention was developed on the basis of these discoveries and provides an anisotropic magnet powder material having the recrystallized fine aggregate structure of an $R_2T_{14}B$ -type intermetallic compound phase, which comprises any of the rare-earth elements including Y (herein referred to as "R"), Fe or a component in which part of the Fe is substituted by Co (herein referred to as "T"), and B as the main components and which components have an average recrystallized grain size of from 0.05 to 20 μm the powder comprising one or more of Si, Ga, Zr, Nb, Mo, Hf, Ta, W, Al, Ti and V (herein referred as "M") characterised in that the material contains a total amount of 0.001 to 5.0 atomic percent of M and the average grain size of the powder is within the range from 5 to 200 μm .

In the R-T-B-M anisotropic magnet powder material according to the invention the powder should have an average grain size of from 5 to 200 μm because an average grain size of under 5 μm is not desirable as it leads to a lower iH_c of bond magnets and full-density magnets made and an average grain size of over 200 μm results on the other hand in a lower magnetic anisotropy in such magnets.

In the R-T-B-M anisotropic magnet material powder of the present invention, part of Fe may be substituted by Cr, Mn, Ni, Cu or Zn, and part of B may be substituted by C, N or O.

EXAMPLES**EXAMPLE 1**

Using a high-frequency melting furnace, an alloy having a chemical composition comprising 11.6% Nd, 0.5% Pr, 11.8% Co, 6.5% B, 0.1% Zr and the balance Fe (atomic %) was melted in an Ar gas atmosphere, and cast into an ingot. This ingot was homogenized in an Ar atmosphere by holding it at a temperature of 1,130°C for 30 hours, and

then crushed to blocks each having a side of up to 20 mm. The block was caused to occlude hydrogen by heating it from room temperature to 750° C in an hydrogen atmosphere under 1 atm. Hydrogen occlusion was caused by holding the block at 750°C for one hour while maintaining the hydrogen atmosphere of 1 atm. to accelerate phase transformation. The block was further heated to 850°C. held at 850°C for one hour, and was forcedly caused to release hydrogen until a 1×10^{-1} vacuum atmosphere was achieved to accelerate phase transformation. The block was then cooled in Ar gas.

The ingot after hydrogen occlusion and release had a recrystallized fine aggregate structure of the $R_2T_{14}B$ -type intermetallic compound phase having an average recrystallization grain size of 0.2 μm . By crushing this ingot to the powder grain sizes as shown in Table 1, samples of an anisotropic magnet powder material of the present invention (hereinafter referred to as "samples of the invention") Nos. 1 to 7 and comparative samples of anisotropic magnet material powder (hereinafter referred to as "comparative samples") Nos. 1 and 2 were prepared.

Each of these samples of the invention Nos. 1 to 7 and comparative samples Nos. 1 and 2 was mixed with 2.5 wt. % epoxy resin and compression-formed while adjusting the density to 6.0 g/cm³ in a magnetic field of 15 kOe to prepare pressurized powder. This pressurized powder was thermoset at 150°C for one hour to prepare an anisotropic bonded magnet. The magnetic properties of the anisotropic magnets prepared are shown in Table 1.

Furthermore, each of these samples of the invention Nos. 1 to 7 and comparative samples Nos. 1 and 2 was compression-formed in a magnetic field into pressurized powder. This pressurized powder was set on a hot press to conduct hot pressing in vacuum at 790°C for ten minutes under a pressure of 1 ton/cm² so that the direction of application of the magnetic field agreed with the direction of compression, and rapidly cooled in Ar gas to prepare an anisotropic full-density magnet. The magnetic properties of the resulting anisotropic full-density magnets are shown in Table 1.

Table 1

Magnet powder	Powder size		Anisotropic bond magnet			Anisotropic full-density magnet		
	Average grain size	(μm)	Br	iHc	BHmax	Br	iHc	BHmax
Invention	1	5	11.1	10.3	23.5	14.6	10.8	43.7
	2	10	11.2	10.4	25.7	14.8	10.5	46.2
	3	50	11.2	10.6	26.5	14.7	10.8	47.4
	4	80	11.2	10.6	27.0	14.7	10.6	47.0
	5	100	11.1	10.7	26.3	14.6	11.0	46.3
	6	150	11.0	10.6	25.1	14.3	10.8	43.9
	7	200	10.8	10.6	23.8	14.2	10.8	43.0
Comparative	1	* 3	10.2	7.6	17.6	14.3	6.1	24.0
	2	* 300	9.6	10.7	18.3	12.4	10.8	34.2

(*: Figures outside the scope of the present invention)

The results shown in Table 1 reveal that the bonded magnets manufactured from the samples of the invention Nos. 1 to 7 having an average grain size within a range of from 5 to 200 μm show better magnetic properties than those of the bonded magnets manufactured from the comparative samples Nos. 1 and 2 having an average grain size outside the range of from 5 to 200 μm.

EXAMPLE 2

Using a high-frequency melting furnace, an alloy having a chemical composition comprising 12.2% Nd, 17.2% Co, 7.0% B, 0.1% Zr, 0.5% Ga and the balance Fe (atomic %) was melted in an Ar gas atmosphere, and cast into an ingot. This ingot was homogenized in an Ar atmosphere by holding it at a temperature of 1,120°C for 40 hours, and then crushed to blocks each having a side of up to 10 mm. The block was caused to occlude hydrogen by heating it from the room temperature to 760°C in a hydrogen atmosphere under 1 atm. Hydrogen occlusion was caused by holding the block at 760°C for one hour while maintaining the hydrogen atmosphere of 1 atm. to accelerate phase transformation. The block was further heated to 820°C, held at 820°C for one hour, and was forcedly caused to release hydrogen until a 1×10^{-1} vacuum atmosphere is achieved to accelerate phase transformation. The block was then cooled in Ar gas.

The ingot after hydrogen occlusion and release had a recrystallized fine aggregate structure of the $R_2T_{14}B$ -type intermetallic compound phase having an average recrystallization grain size of 0.3 μm . By crushing this ingot to an average grain size of 50 μm and 150 μm , samples of the magnet powder of the present invention ion Nos. 8 and 9 were prepared. By crushing this ingot to an average grain size of 300 μm , a sample of comparative magnet powder No. 3 was prepared. The prepared samples of the invention Nos. 8 and 9 had a coercive force, iH_c , of 14.2 kOe, and the comparative sample No. 3 had a coercive force, iH_c , of 14.6 kOe.

Each of the samples of the invention Nos. 8 and 9 and the comparative sample No. 3 was mixed with 2.7 wt.% epoxy resin and compression-formed while making adjustment so as to give a density of 6.0 g/cm³ in an oriented magnetic field in to a pressurized powder. This pressurized powder was thermoset at 130°C for one hour to prepare an anisotropic bonded magnet. The magnetic properties of the prepared anisotropic bonded magnets are represented in a graph as shown in Fig. 1, with H_F/iH_c (where H_F is the oriented magnetic field during forming in the magnetic field; and iH_c is the coercive force of the powder) on the abscissa, and B_r/B_{r70} (where B_r is the remanent magnetization; and B_{r70} is the remanent magnetization in a magnetized field of 70 kOe) on the ordinate.

Furthermore, each of these samples of the invention Nos. 8 and 9 and the comparative sample No. 3 was compression-formed in an oriented magnetic field into pressurized powder. The pressurized powder was set on a hot press and hot-pressed under vacuum at 800°C for ten minutes under a pressure of 1 ton/cm² so that the direction of application of the magnetic field agreed with the direction of compression. The hot-pressed powder was then rapidly cooled in Ar gas to prepare an anisotropic full-density magnet. The magnetic properties of the prepared anisotropic full-density magnet are represented in a graph as shown in Fig. 2, with H_F/iH_c on the abscissa and B_r/B_{r70} on the ordinate.

The results shown in Figs. 1 and 2 suggest that use of the samples of the invention Nos. 8 and 9 having an average grain size of 50 μm and 150 μm , respectively, improves the degree of orientation in a low-orientation magnetic field having an iH_c of up to 1.5 times and permits preparation of an anisotropic bond magnet and an anisotropic full-density magnet having sufficiently high properties, whereas use of the comparative sample No. 3 having an average grain size of 300 μm does not improve the degree of orientation in an oriented magnetic field having an iH_c of up to 1.5 times, and does not give an anisotropic bonded magnet or an anisotropic full-density magnet having sufficiently high properties.

According to the rare-earth magnet material powder excellent in anisotropy of the present invention having an average grain size within a range of from 5 to 200 μm , the degree of orientation in a low-orientation magnetic field of a coercive force, iH_c , of up to 1.5 times is improved, and it is possible to manufacture an anisotropic rare-earth magnet having better magnetic properties than those of conventional ones in a low magnetic field output, thus providing industrially useful effects.

Claims

1. An anisotropic magnet powder material having the recrystallized fine aggregate structure of an $R_2T_{14}B$ -type intermetallic compound phase, which comprises any of the rare-earth elements including Y, herein referred to as "R", Fe or a component in which part of the Fe is substituted by Co, herein referred to as "T", and B as the main components and which components have an average recrystallized grain size of from 0.05 to 20 μm , said powder further comprising one or more of Si, Ga, Zr, Nb, Mo, Hf, Ta, W, Al, Ti and V, herein referred as "M" characterised in that the material contains a total amount of 0.001 to 5.0 atomic percent of M and the average grain size of the powder is within the range from 5 to 200 μm .
2. A powder material as claimed in Claim 1 wherein part of the Fe is additionally or alternatively substituted by Cr, Mn, Ni, Cu or Zn.
3. A powder material as claimed in Claim 1 or Claim 2 wherein part of the B is substituted by C, N or O.

4. Use of the powder as claimed in any one of the preceding claims in the manufacture of bonded or hot-pressed magnets.

5 Patentansprüche

1. Anisotropes Magnetpulvermaterial mit der rekristallisierten feinen Aggregatstruktur einer intermetallischen Verbindungsphase des Typs $R_2T_{14}B$, welche eines der Seltenerdelemente, einschließlich Y, hierin als "R" bezeichnet, Fe oder einen Bestandteil, in dem ein Teil des Fe durch Co substituiert ist, hierin als "T" bezeichnet, und B als die Hauptbestandteile umfaßt und wobei die Bestandteile eine durchschnittliche rekristallisierte Korngröße von 0,05 bis 20 μm aufweisen, wobei das Pulver weiterhin eines oder mehrere von Si, Ga, Zr, Nb, Mo, Hf, Ta, W, Al, Ti und V, hierin als "M" bezeichnet, umfaßt, dadurch gekennzeichnet, daß das Material eine Gesamtmenge von 0,001 bis 5,0 Atomprozent M enthält und daß die durchschnittliche Korngröße des Pulvers innerhalb des Bereichs von 5 bis 200 μm liegt.
2. Pulvermaterial nach Anspruch 1, worin ein Teil des Fe zusätzlich oder alternativ durch Cr, Mn, Ni, Cu oder Zn substituiert ist.
3. Pulvermaterial nach Anspruch 1 oder 2, worin ein Teil des B durch C, N oder O substituiert ist.
4. Verwendung des Pulvers nach einem der vorhergehenden Ansprüche bei der Herstellung von gebundenen oder heißgepreßten Magneten.

25 Revendications

1. Matériau en poudre anisotrope pour aimant, ayant la structure globale fine recristallisée d'une phase de composé intermétallique du type $R_2T_{14}B$, qui comprend l'un quelconque des éléments des terres rares y compris Y, désigné ci-après par le symbole "R", du fer ou un composant dans lequel une partie du fer est remplacée par du cobalt, désigné ci-après par le symbole "T", et du bore comme composants principaux, ces composants ayant un diamètre de grain moyen à l'état recristallisé de 0,05 à 20 μm , cette poudre comprenant en outre l'un ou plusieurs des éléments Si, Ga, Zr, Nb, Mo, Hf, Ta, W, Al, Ti et V, désignés ci-après par le symbole "M", matériau caractérisé en ce qu'il contient une quantité totale de 0,001 à 5,0 pour cent, en proportion atomique, de M et en ce que la poudre a un diamètre des grains moyens dans la plage de 5 à 200 μm .
2. Matériau en poudre suivant la revendication 1, dans lequel une partie du fer est remplacée en outre ou en variante par du chrome, du manganèse, du nickel, du cuivre ou du zinc.
3. Matériau en poudre suivant la revendication 1 ou la revendication 2, dans lequel une partie du bore est remplacée par du carbone, de l'azote ou de l'oxygène.
4. Utilisation de la poudre suivant l'une quelconque des revendications précédentes dans la fabrication d'aimants liés ou pressés à chaud.

Figure 1

Magnetic properties of anisotropic bonded magnets

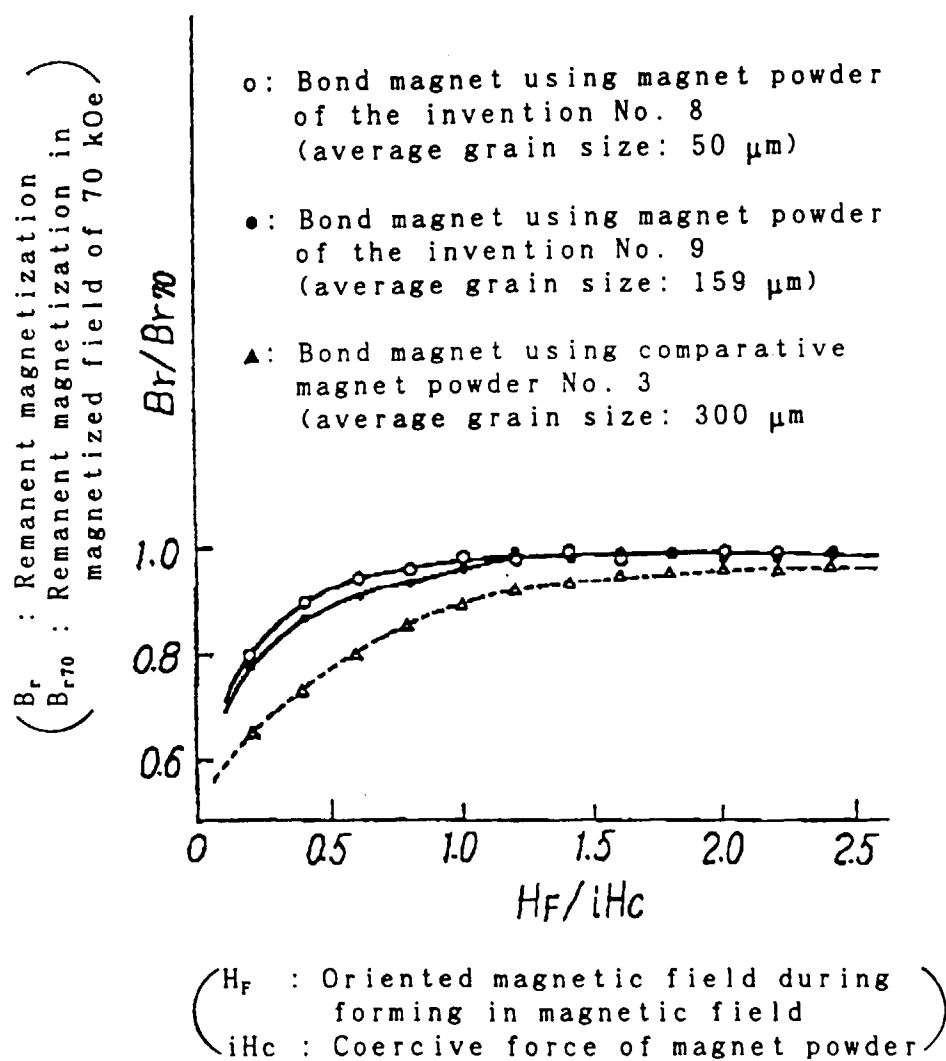


Figure 2

Magnetic properties of anisotropic full-density magnets

