⑲ Europäisches Patentamt
European Patent Office
Office européen des brevets

⑪ Publication number: **0 289 680 B1**

⑫ EUROPEAN PATENT SPECIFICATION

㊺ Date of publication of patent specification: **22.06.94** (51) Int. Cl.^{5}: **H01F 1/053**, H01F 41/02

㉑ Application number: **87308666.4**

㉒ Date of filing: **30.09.87**

(54) Permanent magnet and method of producing the same.

㉚ Priority: **30.04.87 JP 104623/87**

㊸ Date of publication of application:
09.11.88 Bulletin 88/45

㊺ Publication of the grant of the patent:
22.06.94 Bulletin 94/25

(84) Designated Contracting States:
AT CH DE FR GB IT LI NL

(56) References cited:
EP-A- 0 175 214
FR-A- 2 586 323

PATENT ABSTRACTS OF JAPAN, vol. 11, no. 232 (C-437)[2679], 29th July 1987; & JP-A-62 47 455 (SUMITOMO SPECIAL METALS CO.) 02-03-1987

JOURNAL OF APPLIED PHYSICS, vol. 57, no. 8, part 2B, April 1985, pages 4106-4108; E.B. BOLTICH et al.: "Magnetic characteristics of R2Fe14B systems prepared with high purity rare earths (R = Ce,Pr,Dy, and Er)"

JOURNAL DE PHYSIOUE, COLLOOUE C8, Supplément au n 12, Tome 49, décembre 1988, pages 631-632

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Description

The present invention relates to a permanent magnet which comprises a rare earth element, iron and boron, and a method of producing the same.

The term "rare earth element" (R) is used in this specification in a wide sense as including Yttrium (Y).

A permanent magnet is one of the major components used in the electrical and electronic field, e.g. in various household electrical appliances and in the peripheral console units of large computers.

With a recently growing demand for smaller and higher-efficiency electrical appliances, permanent magnets have increasingly been required to have a higher and higher performance. Typical permanent magnets now in use include an alnico hard ferrite magnet and a rare earth element -transition metal magnet. In particular, an R - Co permanent magnet and an R - Fe - B permanent magnet, which are rare earth element - transition metal magnets, can produce a high magnetic performance, so that many researches have hitherto been made on them.

For example, the following methods have been devised for manufacturing an R -Fe - B permanent magnet.

(1) A method of sintering a material on the basis of a powder metallurgy technique (References 1 and 2, referred to below).

(2) A resin bonding method using rapidly-quenched thin fragments obtained by a melt spinning method. A rapidly-quenched thin fragment of about 30 µm thick is produced by a melt spinning apparatus which is used for manufacturing an amorphous alloy, and a magnet is produced from the thin fragments by adopting a resin bonding method. (References 3 and 4, referred to below).

(3) A method of mechanically aligning the rapidly-quenched thin fragments used in method (2) by a two-stage hot pressing method (References 4 and 5, referred to below).

Reference 1: Japanese Patent Laid-Open Specification No. 46008/1984.

Reference 2: M. Sagawa, S. Fujimura, N. Togawa, H. Yamamoto, and Y Matushita: J. Apl. Phys. Vol, 55 (6), 15th March p. 2083 (1984).

Reference 3: Japanese Patent Laid-Open Specification No. 211549/1984.

Reference 4: R.W. Lee: Appl. Phys. Lett. Vol. 46(8), 15th April p. 790 (1985).

Reference 5: Japanese Patent Laid-Open Specification No. 100402/1985.

In the sintering method (1), an alloy ingot is first made by melting and casting, and is pulverized to a particle diameter of about 3 µm. The pulverized powder is kneaded with a binder, and pressed in a magnetic field to obtain a moulded body.

The moulded body is sintered at approximately 1,100 °C in an argon gas atmosphere for 1 hour, and thereafter heat treated at approximately 600 °C to improve the coercive force.

In the resin bonding method (2) which incorporates a melt spinning method, rapidly-quenched thin fragments of an R - Fe - B alloy are first produced by a melt spinning apparatus at an optimum substrate velocity. A thus-obtained ribbon-like thin fragment having a thickness of 30 µm is an aggregate of crystal grains having a diameter of not more than 1,000Å. Since the crystal axes of the crystal grains are distributed isotropically, the thin ribbon is magnetically isotropic. If the thin ribbon is pulverized into an appropriate grain size, kneaded with a resin, and then pressed, an isotropic magnet is obtained.

In the manufacturing method (3) which utilises a two-stage hot pressing process, the ribbon-like thin fragments used in the method (2) are pressed at a temperature of about 700 °C and under a pressure of not more than 1.4 ton/cm^2 in vacuo or in an inert gas. Then this pressed body is next pressed at 700 °C and under a pressure of 7 ton/cm^2 for several seconds to reduce the thickness to half the initial thickness. Thus, a dense and anisotropic R - Fe - B magnet is obtained.

Furthermore, a liquid dynamic compaction process (hereinunder referred to as "LDC" process) is also used for producing an alloy having a coercive force in a bulk state (Reference 6, referred to below).

Reference 6: T.S. Chin et al: J. Appl. Phys. 59 (4), 15th February, p. 1297 (1986).

Although a permanent magnet containing a rare earth element, iron and boron as the basic ingredients can be made by the methods of the prior art, these methods have the defects referred to below.

The sintering process (1) necessitates the step of powdering an alloy. Since the powder of an R - Fe - B alloy is very reactive to oxygen, it is necessary to handle the charge of the powder used in the sintering process very carefully and an expensive equipment for inert gas, etc. is required.

In addition, in the sintering process,the carbon of a binder has a deleterious influence on the magnetic performance, and is difficult to handle the moulded body called a green body. These problems lower the productivity, and it cannot therefore be said that the method (1) is capable of making the best use of an R - Fe - B magnet whose main advantage is that it is inexpensive in the material cost.

Both methods (2) and (3) require an expensive vacuum melt spinning apparatus or hot press which has a poor productivity.

A magnet produced by the method (2) is isotropic and, hence, it is impossible to obtain a high energy product. This magnet is therefore disadvantageous both in its temperature characteristics and in use.

The method (3) uses a two-stage hot pressing process. Therefore, the productivity is very poor, and it cannot make the best use of an R - Fe - B magnet which, as stated above, is inexpensive in its material cost.

The LDC process also has the problems of requiring an expensive equipment and having a poor productivity.

Accordingly, it is an object of the present invention to eliminate the above-described problems in the prior art and to provide a high-performance and low-cost rare earth-iron-boron permanent magnet and a method of manufacturing the same.

According, therefore, to the present invention, there is provided a permanent magnet as recited in Claim 1.

Preferably, the alloy contains Nd and/or Pr.

The invention also comprises a method of making a permanent magnet as recited in Claim 9.

In a first embodiment of the said method, the magnet is heat treated at a temperature not lower than 250 °C.

In a second embodiment of the said method, the magnet is subjected to hot processing at a temperature not lower than 500 °C so as to make the magnet anisotropic.

In a third embodiment of the said method, the method comprises hot processing the magnet at a temperature not lower than 500 °C so as to make the magnet anisotropic, and heat treating the magnet at a temperature not lower than 250 °C.

A preferred composition of a permanent magnet containing at least one rare earth element, iron and boron as basic ingredients is 8 to 30 atm% of a rare earth element or elements, 2 to 28 atm% of boron, the balance being substantially iron.

As the rare earth element or elements employed Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu are usable. Above all, Nd and Pr are preferable.

Two or more of these rare earth elements may be used in combination. In addition to these basic ingredients, impurities inevitable in the manufacturing process may be contained in the alloy, and cobalt may be added, e.g. in an amount of up to 40 atm%, in order to raise the Curie temperature. Moreover, Aℓ, Cr, Mo, W, Nb, Ta, Zr, Hf, Ti, etc. e.g. in an amount of up to10 atm%, may be added in order to enhance the coercive force.

As indicated above, the carbon content and the oxygen content in the magnet are set at no more than 400 ppm and 1,000 ppm, respectively, because if they exceed 400 ppm and 1,000 ppm, respectively, the magnetic performance is lowered.

In order to obtain a desired coercive force in a bulk state in an R - Fe - B magnet, the grain diameter of the crystal grains must be appropriate.

If the average grain diameter of the magnet after casting exceeds 150 µm, the coercive force does not reach that of a ferrite magnet, namely 4 KOe, even after hot processing, and such a R - Fe - B alloy cannot be said to be a practical permanent magnet alloy. Therefore, the average grain diameter must be not more than 150 µm. The grain diameter can be controlled by varying the cooling temperature by altering the material of a mould, the heat capacity of the mould, etc.

A heat treatment after casting is necessary for diffusing the Fe phase which exists as the primary crystal in the cast alloy, thereby elminating a magnetically soft phase. It goes without saying that a similar heat treatment carried out after hot processing is effective for improving the magnetic properties.

The hot processing at a temperature of not lower than 500 °C is effective for orientating the crystal axes of the crystal grains to make the magnet anisotropic and for making the crystal grains finer, thereby greatly enhancing the magnetic properties.

The invention is illustrated, merely by way of example, in the accompanying drawing which is a graph showing the relationship between the average grain diameter (µm) of the crystals after casting and the coercive force (iHc) after hot pressing in an embodiment of the present invention.

Example 1

Table 1 below shows the compositions, in atm%, of permanent magnets containing various rare earth elements, iron, boron as the basic ingredients which were produced in the following procedure.

An alloy having a desired composition was melted in an Ar atmosphere in an induction furnace and cast into various moulds at 1,000 ° C. When 20 minutes had passed after casting the ingots were taken out. The alloy contained a rare earth metal having a purity of 95% (the impurities being mainly other rare earth metals), and the alloy contained a transition metal having a purity of not less than 99.9%. As boron, a ferroboron alloy was used.

The cast alloy was subjected to heat treatment at a temperature of not lower than 250 ° C (in Example 1, at 1,000 ° C for 24 hours), and was then cut to obtain a permanent magnet.

The magnetic performance and the average grain diameter of the magnet obtained by casting each composition into an iron mould is shown in Table 2 below.

The accompanying drawing shows the relationship between the average diameter (μm) after casting and the coercive force (iHc) after hot pressing of the samples Nos. 3 and 4 having the respective compositions shown in Table 1. The grain diameter was controlled by using a water-cooled copper mould, an iron mould, a ceramic mould, etc, and by vibrating the mould. From this result, it is found that it is possible to obtain a permanent magnet by casting while controlling the grain diameter.

Table 1

Sample No.	Composition (atm %)					
1	Nd_{15}	Fe_{77}	B_8			
2	Nd_{15}	Fe_{80}	B_5			
3	Pr_{16}	Fe_{80}	B_4			
4	Pr_{16}	$Fe_{81.5}$	$B_{2.5}$			
5'	Pr_{17}	Fe_{77}	B_6			
6	Ce_2	Nd_5	Pr_{10}	Fe_{79}	B_4	
7	Nd_{10}	Pr_7	Fe_{70}	Co_5	B_8	
8	Nd_5	Pr_{12}	Fe_{76}	Al_3	B_4	
9	Nd_{20}	Dy_2	Fe_{70}	Co_2	B_6	
10	Pr_{10}	Tb_2	Fe_{74}	Co_2	Al_2	B_{10}

Table 2

Sample No.	IHc (KOe)*	Average grain Diameter (μm)
1	5.1	100
2	5.7	80
3	7.7	30
4	6.5	23
5	6.3	65
6	7.3	33
7	5.9	67
8	8.0	28
9	4.4	47
10	1.1	150

* 1 De = 79,6 $\frac{A}{m}$;

Example 2

The compositions shown in Table 3 below were cast into a water cooled copper mould in the same way as in Example 1, and thereafter the ingots were hot pressed at 1,000 ° C to make the respective permanent magnets anisotropic.

The average diameter and the magnetic performance after heat treatment, and the average diameter and the magnetic performance after hot pressing, of each magnet are shown in Table 4 below.

The magnetic properties of the samples Nos. 11, 13 and 14 after hot pressing and 24-hour heat treatment carried out at 1,000 ° C after the hot pressing are shown in Table 5 below.

Table 3

Sample No.	Composition (atm %)
11	Pr_{17} Fe_{79} B_4
12	Pr_{14} Dy_2 Fe_{79} B_5
13	Pr_{13} Nd_4 Fe_{74} Co_5 B_4
14	Pr_{16} Fe_{70} Co_5 Al_3 B_6
15	Nd_{13} Tb_2 Fe_{66} Co_{10} Al_5 B_4
16	Ce_2 Pr_{13} Nd_2 Fe_{71} Co_5 Cr_1 Zr_1 Ti_1 B_4

Table 4

	After Casting			After Hot Pressing		
Sample No.	Average Grain Diameter (µm)	iHc (KOe)	(BH)max (MGOe) *	Average Grain Diameter (µm)	iHc (KOe)	(BH)max (MGOe)
11	15	8.8	5.8	10	10.5	24.6
12	30	7.7	4.8	20	8.8	21.3
13	23	8.0	5.5	13	9.0	23.8
14	40	6.7	4.7	28	7.0	20.2
15	75	5.8	3.1	45	6.8	18.5
16	20	8.0	5.3	10	9.7	21.4

Table 5

Sample No.	Average Grain Diameter (µm)	iHc (KOe)	Br (KG) **	(BH)max (MGOe)
11	10	11.0	11.0	25.1
13	13	9.5	10.4	24.3
14	28	8.0	10.2	22.4

* 1 MGOe = 7,96 $\frac{kJ}{m^3}$;
** 1 G = 10^{-4}T;

As is obvious from the above results, the hot pressing makes the grain diameters smaller and greatly enhances the magnetic performance, and the heat treatment improves the magnetic performance.

In the embodiments of the present invention described above, the casting method was adopted,and the carbon content and the oxygen content in the magnet obtained were not more than 400 ppm and not more than 1,000 ppm, respectively.

As described above, since the permanent magnets of the present invention can be produced in bulk with a satisfactory coercive force without the need for pulverizing a cast ingot, it is possible to greatly simplify the manufacturing steps, and a high-performance and low-cost permanent magnet can be obtained.

Claims

1. A permanet magnet cosisting of a body cast from an alloy having at least one rare earth element, iron and boron characterised in that the average grain diameter of the crystals of the magnet is not more than 150 µm and the carbon and oxygen content of the magnet are not more than 400 ppm and 1,000 ppm, respectively.

2. A permanent magnet as claimed in claim 1 characterised in that the alloy contains Nd and/or Pr.

3. A permanent magnet as claimed in claim 1 or 2 characterised in that the alloy contains 8 to 30 at% of a rare earth element or elements and 2 to 28 at% of boron, the balance being at least mainly iron.

4. A permanent magnet as claimed in claim 3 characterised in that the said balance is made up wholly of iron and impurities.

5. A permanent magnet as claimed in claim 3 characterised in that the said balance contains cobalt.

6. A permanent magnet as claimed in claim 5 characterised in that the cobalt content of the alloy does not exceed 40 at%.

7. A permanent magnet as claimed in any of claims 3, 5 or 6 characterised in that the said balance contains one or more of the elements Aℓ, Cr, Mo, W, Nb, Ta, Zr, Hf andTi.

8. A permanent magnet as claimed in claim 7 characterised in that the said balance contains not more than 10 at% of the said one or more elements.

9. A method of making a permanent magnet comprising the steps of:-
casting a molten alloy comprising at least one rare earth element, iron and boron to produce a cast body and further processing the cast body to produce the permanent magnet characterised in that the average grain diameter of the crystals of the magnet is not more than 150 µm and the carbon and oxygen content of the magnet are not more than 400 ppm and 1,000 ppm respectively.

10. A method as claimed in claim 9 characterised in that, after the alloy has been cast, it is heat treated at a temperature not lower than 250 ° C.

11. A method as claimed in claim 9 or 10, characterised in that the cast alloy is subjected to hot processing at a temperature not lower than 500 ° C so as to make the magnet anisotropic.

12. A method as claimed in any of Claims 9 to 11, characterised in that the said casting is conducted in an Ar atmosphere.

13. A method as claimed in any of Claims 9 to 12, characterised in that said molten material is cast into a water cooled copper mould.

Patentansprüche

1. Permanentmagnet, bestehend aus einem Körper, der gegossen ist aus mindestens einem Seltenerdelement, Eisen und Bor,
dadurch gekennzeichnet,
daß der mittlere Korndurchmesser der Kristalle des Magneten nicht mehr als 150 µm ist und der Kohlenstoff- und Sauerstoffgehalt des Magneten nicht mehr als 400 ppm bzw. 1.000 ppm ist.

2. Permanentmagnet nach Anspruche 1,
dadurch gekennzeichnet,
daß die Legierung Nd und/oder Pr enthält.

3. Permanentmagnet nach Anspruch 1 oder 2,
dadurch gekennzeichnet,
daß die Legierung 8 bis 30 Atom-% eines Seltenerdelements oder von Seltenerdelementen und 2 bis 28 Atom-% Bor enthält, wobei der Rest zumindest hauptsächlich Eisen ist.

4. Permanentmagnet nach Anspruch 3,
dadurch gekennzeichnet,
daß der Rest ganz aus Eisen und Verunreinigungen besteht.

5. Permanentmagnet nach Anspruch 3,
dadurch gekennzeichnet,
daß der Rest Kobalt enthält.

6. Permanentmagnet nach Anspruch 5,
dadurch gekennzeichnet,
daß der Kobalt-Gehalt der Legierung 40 Atom-% nicht überschreitet.

7. Permanentmagnet nach einem der Ansprüche 3, 5 oder 6,
dadurch gekennzeichnet,
daß der Rest eines oder mehrere der Elemente Al, Cr, Mo, W, Nb, Ta, Zr, Hf und Ti enthält.

8. Permanentmagnet nach Anspruch 7,
dadurch gekennzeichnet,
daß der Rest nicht mehr als 10 Atom-% des einen oder der mehreren Elemente enthält.

9. Verfahren zur Herstellung eines Permanentmagneten, folgende Schritte aufweisend:
Gießen eines geschmolzenen Materials, das mindestens ein Seltenerdelement, Eisen und Bor enthält, zur Erzeugung eines Gußkörpers, und weiteres Behandeln des Gußkörpers zur Erzeugung des Permanentmagneten,
dadurch gekennzeichnet,
daß der mittlere Korndurchmesser der Kristalle des Magneten nicht mehr als 150 µm beträgt und der Kohlenstoff- und Sauerstoffgehalt des Magneten nicht mehr als 400 ppm bzw. 1.000 ppm beträgt.

10. Verfahren nach Anspruch 9,
dadurch gekennzeichnet,
daß die Legierung, nachdem sie gegossen wurde, bei einer Temperatur nicht unterhalb von 250 °C wärmebehandelt wird.

11. Verfahren nach Anspruch 9 oder 10,
dadurch gekennzeichnet,
daß die gegossene Legierung bei einer Temperatur nicht unterhalb von 500 °C einer Heißbehandlung unterzogen wird, um den Magneten anisotrop zu machen.

12. Verfahren nach einem der Ansprüche 9 bis 11,
dadurch gekennzeichnet,
daß das Gießen in einer Ar-Atmosphäre durchgeführt wird.

13. Verfahren nach einem der Ansprüche 9 bis 12,
dadurch gekennzeichnet,
daß das geschmolzene Material in eine wassergekühlte Kupferkokille gegossen wird.

Revendications

1. Aimant permanent constitué d'un corps coulé à partir d'au moins un élément des terres rares, le fer et le bore, caractérisé en ce que le diamètre moyen des grains des cristaux de l'aimant n'est pas supérieur à 150 µm, et que la teneur de l'aimant en carbone et en oxygène n'est pas supérieure respectivement à 400 et 1 000 ppm.

2. Aimant permanent selon la revendication 1, caractérisé en ce que l'alliage contient Nd et/ou Pr.

3. Aimant permanent selon la revendication 1 ou 2, caractérisé en de que l'alliage contient de 8 à 30 %-atm d'un ou plusieurs éléments des terres rares et de 2 à 28 %-atm de bore, le reste étant constitué au moins essentiellement de fer.

4. Aimant permanent selon la revendication 3, caractérisé en ce que ledit reste est constitué en totalité de fer et d'impuretés.

5. Aimant permanent selon la revendication 3, caractérisé en ce que ledit reste est constitué de cobalt.

6. Aimant permanent selon la revendication 5, caractérisé en ce que la teneur de l'alliage en cobalt ne dépasse pas 40 %-atm.

7. Aimant permanent selon l'une quelconque des revendications 3, 5 ou 6, caractérisé en ce que ledit reste contient un ou plusieurs des éléments A, Cr, Mo, W, Nb, Ta, Zr, Hf et Ti.

8. Aimant permanent selon la revendication 7, caractérisé en ce que ledit reste contient au plus 10 %-atm de l'un ou plusieurs desdits éléments.

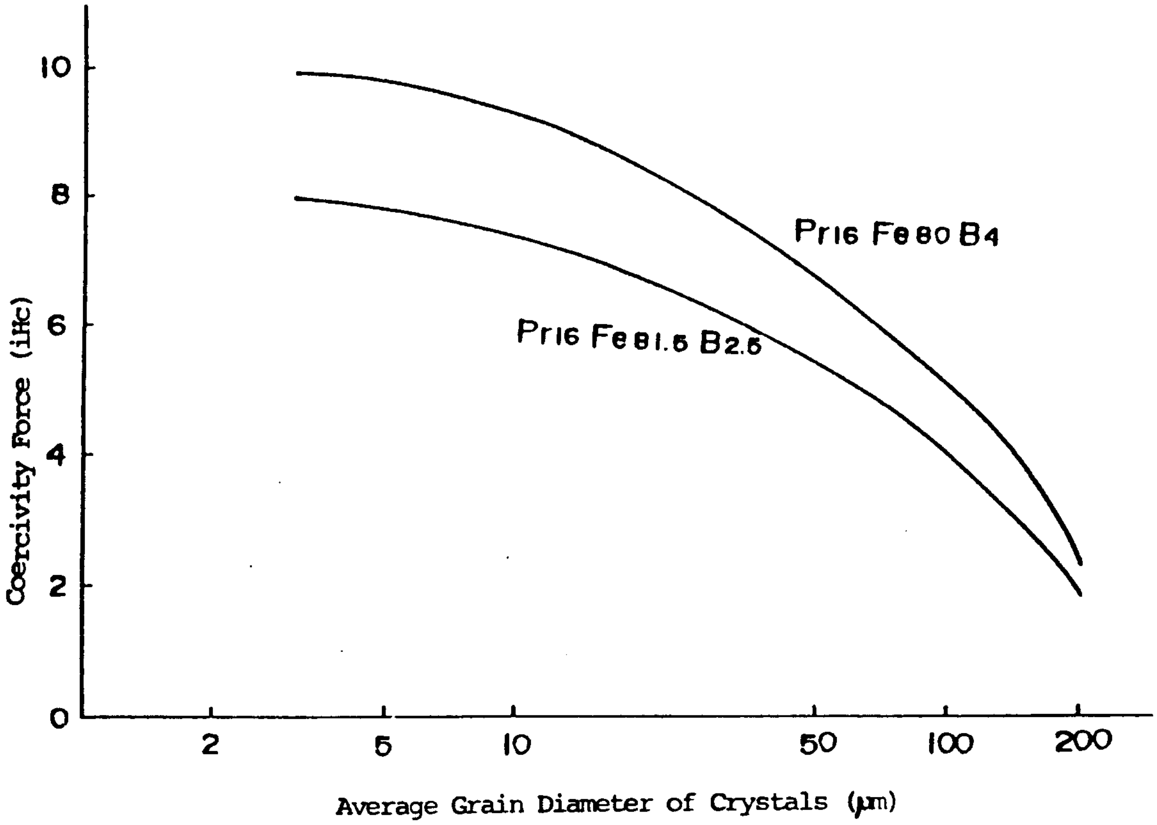
9. Procédé pour fabriquer un aimant permanent, qui comprend les étapes consistant à couler un matériau fondu comprenant au moins un élément des terres rares, du fer et du bore, pour produire un corps coulé, puis à mettre en oeuvre le corps coulé pour produire un aimant permanent, caractérisé en ce que le diamètre moyen des grains des cristaux de l'aimant n'est pas supérieur à 150 µm, et que la teneur de l'aimant en carbone et en oxygène n'est pas supérieure respectivement à 400 et 1 000 ppm.

10. Procédé selon la revendication 9, caractérisé en ce que, après coulée de l'alliage, on le soumet à un traitement thermique à une température non inférieure à 250 °C.

11. Procédé selon la revendication 9 ou 10, caractérisé en ce que l'alliage coulé est soumis à une transformation à chaud à une température non inférieure à 500 °C pour rendre l'aimant anisotrope.

12. Procédé selon l'une quelconque des revendications 9 à 11, caractérisé en ce que la coulée est effectuée dans une atmosphère de Ar.

13. Procédé selon l'une quelconque des revendications 9 à 12, caractérisé en ce que le matériau fondu est coulé dans un moule en cuivre refroidi à l'eau.


Coercivity Force (iHc)
0
2
4
6
8
10
2
5
10
50
100
200
Average Grain Diameter of Crystals (μm)
Pr16 Fe80 B4
Pr16 Fe81.5 B2.5