



Europäisches Patentamt
European Patent Office
Office européen des brevets

Publication number:

**0 289 680
A2**

12

EUROPEAN PATENT APPLICATION

21 Application number: 87308666.4

51 Int. Cl.4: **H01F 1/04 , H01F 41/02**

22 Date of filing: 30.09.87

30 Priority: 30.04.87 JP 104623/87

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

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

71 Applicant: **SEIKO EPSON CORPORATION**
4-1, Nishishinjuku 2-chome
Shinjuku-ku Tokyo-to(JP)

72 Inventor: **Akioka, Koji**
c/o SEIKO EPSON CORPORATION 3-5, Owa
3-chome
Suwa-shi Nagano-ken(JP)
Inventor: **Kobayashi, Osamu**
c/o SEIKO EPSON CORPORATION 3-5, Owa
3-chome
Suwa-shi Nagano-ken(JP)
Inventor: **Shimoda, Tatsuya**
c/o SEIKO EPSON CORPORATION 3-5, Owa
3-chome
Suwa-shi Nagano-ken(JP)

74 Representative: **Miller, Joseph et al**
J. MILLER & CO. Lincoln House 296-302 High
Holborn
London WC1V 7JH(GB)

54 **Permanent magnet and method of producing the same.**

57 A permanent magnet made from an alloy comprising at least one rare earth element, iron and boron characterised in that the average grain diameter of the crystals of the alloy is not more than 150 μm , the carbon content and the oxygen content of the alloy being not more than 400 ppm and not more than 1000 ppm respectively.

EP 0 289 680 A2

PERMANENT MAGNET AND METHOD OF PRODUCING THE SAME

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. Appl. 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/cm² in vacuo or in an inert gas. Then this pressed body is next pressed at 700°C and under a pressure of 7 ton/cm² 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 (hereinafter 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.

5 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.

10 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 made from an alloy comprising at least one rare earth element, iron and boron characterised in that the average grain diameter of the crystals of the alloy is not more than 150 μm , the carbon content and the oxygen content of the alloy being not more than 400 ppm and not more than 1000 ppm respectively.

Preferably, the alloy contains Nd and/or Pr.

The invention also comprises a method of making a permanent magnet comprising casting an alloy comprising at least one rare earth element, iron and boron characterised in that the average grain diameter of the crystals of the alloy is not more than 150 μm , the carbon content and the oxygen content of the alloy being not more than 400 ppm and not more than 1000 ppm respectively.

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

25 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.

30 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.

35 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 to 10 atm%, may be added in order to enhance the coercive force.

40 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.

45 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.

50 A heat treatment after casting is necessary for diffusing the Fe phase which exists as the primary crystal in the cast alloy, thereby eliminating 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.

55 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 ferrobore 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 and ground 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

5

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

30

Example 2

35

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.

40

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.

45

50

55

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	

25

30

35

40

45

50

55

Table 4

Sample No.	After Casting			After Hot Pressing		
	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

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 permanent magnet made from an alloy comprising at least one rare earth element, iron and boron characterised in that the average grain diameter of the crystals of the alloy is not more than 150 μm , the carbon content and the oxygen content of the alloy being not more than 400 ppm and not more than 1000 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 atm% of a rare earth element or elements and 2 to 28 atm% 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
5 not exceed 40 atm%.

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 and Ti.

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

9. A method of making a permanent magnet comprising casting an alloy comprising at least one rare
10 earth element, iron and boron characterised in that the average grain diameter of the crystals of the alloy is not more than 150 μm , the carbon content and the oxygen content of the alloy being not more than 400 ppm and not more than 1000 ppm respectively.

10. A method as claimed in claim 9 characterised in that, after the alloy has been cast, it is heat treated
15 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.

20

25

30

35

40

45

50

55

