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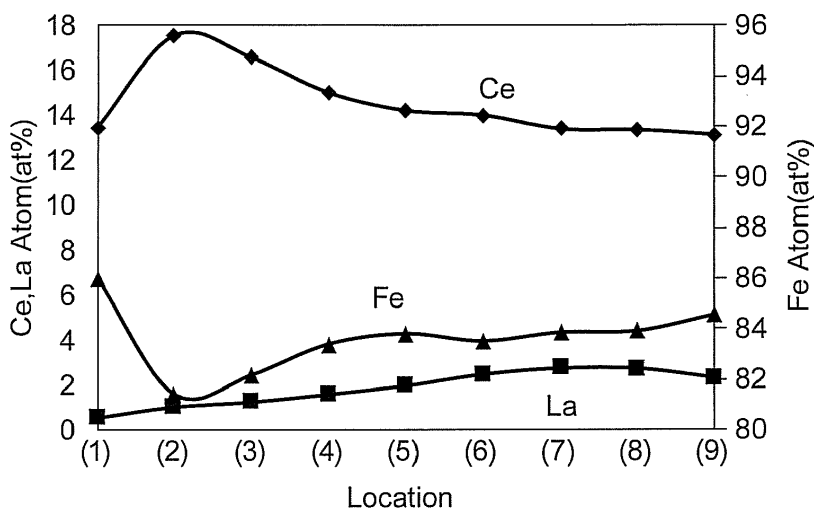
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(54) **RARE EARTH MAGNET**

(57) A rare earth magnet having an overall composition represented by the formula of $(La_xCe_{(1-x)})_yFe_{(100-y-w-z-v)}Co_wB_zM_v$ (wherein M is one or more elements selected from Ga, Al, Cu, Au, Ag, Zn, In and Mn, and an unavoidable impurity elements, and x, y, z, w and v satisfy the following relationship: $0.10 \leq x \leq 0.30$, $5 \leq y \leq 20$, $4 \leq z \leq 6.5$, $0 \leq w \leq 8$, and $0 \leq v \leq 2.0$).

FIG. 5



Description

TECHNICAL FIELD

5 **[0001]** The present invention relates to a rare earth magnet. More specifically, the present invention relates to a (La, Ce)-Fe-B-based rare earth magnet.

BACKGROUND ART

10 **[0002]** An R-Fe-B-based rare earth magnet (R is a rare earth element) has a main phase and a grain boundary phase surrounding the main phase. The main phase has a composition represented by $R_2Fe_{14}B$ and is a magnetic phase. In the following description, the phase having a composition represented by $R_2Fe_{14}B$ is sometimes referred to as " $R_2Fe_{14}B$ phase".

15 **[0003]** In the R-Fe-B-based rare earth magnet, when R is Nd, the magnetic properties are excellent in particular. Accordingly, an Nd-Fe-B-based rare earth magnet is often used as a high-performance magnet, e.g., in a motor for driving an electric vehicle, a hybrid vehicle, etc.

[0004] Patent Document 1 discloses a technique of replacing part of Nd in the Nd-Fe-B-based rare earth magnet by Ce, La and/or Y to improve the hot workability of the rare earth magnet.

20 **[0005]** In the Nd-Fe-B-based rare earth magnet disclosed in Patent Document 1, the magnetic properties are reduced in proportion to the replacement of part of Nd in the $Nd_2Fe_{14}B$ phase by Ce, La and/or Y. Accordingly, hot workability is improved only to an extent that reduction in magnetic properties does not pose a problem in practice. As a result, the replacement ratio of Nd by Ce, La and/or Y is small.

[0006] The price for Nd is sharply increasing, and attempts are made to replace part of Nd in the Nd-Fe-B-based rare earth magnet by Ce, La, Gd, Y and/or Sc which are less expensive than Nd.

25 **[0007]** As an example thereof, Patent Document 2 discloses a rare earth magnet having a main phase and a grain boundary phase surrounding the main phase, where the main phase has a so-called core/shell structure. In Patent Document 2, it is disclosed that the core is a $Ce_2Fe_{14}B$ phase and the shell is a $(Nd_{0.5}Ce_{0.5})Fe_{14}B$ phase. Patent Document 2 also discloses, as the method of producing the rare earth magnet, a method of bringing a green compact or sintered body of a Ce-Fe-B-based rare earth magnet powder into contact with an Nd-Cu alloy and thereby causing

30 Nd in the Nd-Cu alloy melt to diffuse into the Ce-Fe-B-based rare earth magnet powder.

[0008] In an example disclosed in Patent Document 2, the core is a $Ce_2Fe_{14}B$ phase and does not contain Nd. On the other hand, the shell is a $(Nd_{0.5}Ce_{0.5})Fe_{14}B$ phase and contains Nd. Furthermore, the grain boundary surrounding the main phase contains a lot of Nd due to penetration of the Nd-Cu alloy melt.

35 **[0009]** As such, in an example disclosed in Patent Document 2, since the core is magnetically separated by the shell and the grain boundary which are rich in Nd, magnetization inversion is not propagated across a plurality of cores. Accordingly, despite the fact that the core does not contain Nd and the magnetic properties of the core itself are reduced, the magnetization of the rare earth magnet as a whole is not so much decreased. This means that unless the $Ce_2Fe_{14}B$ phase is surrounded by an Nd-rich phase, desired magnetic properties cannot be ensured in the entire rare earth magnet.

40 RELATED ART

 Patent Document

[0010]

45 [Patent Document 1] Japanese Unexamined Patent Publication No. 4-21744
 [Patent Document 2] International Publication No. 2014/196605

SUMMARY OF THE INVENTION

50 Problems to be Solved by the Invention

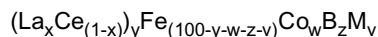
[0011] The present inventors have found a problem that in the R-Fe-B-based rare earth magnet, magnetization of the main phase must be enhanced so as to enhance the magnetic properties of the rare earth magnet as a whole by using, as R, Ce that is less expensive than Nd.

55 **[0012]** The present invention has been made to solve the problem above. An object of the present invention is to provide a rare earth magnet in which magnetization is enhanced without using Nd as R of the R-Fe-B-based rare earth magnet.

Means to Solve the Problems

[0013] As a result of enormous intensive studies to attain the object above, the present inventors have accomplished the present invention. The gist of the present invention is as follows.

<1> A rare earth magnet having an overall composition represented by the formula:



(wherein M is one or more elements selected from Ga, Al, Cu, Au, Ag, Zn, In and Mn, and an unavoidable impurity elements, and

x, y, z, w and v satisfy the following relationship:

$$0.10 \leq x \leq 0.30,$$

$$5 \leq y \leq 20,$$

$$4 \leq z \leq 6.5,$$

$$0 \leq w \leq 8,$$

$$0 \leq v \leq 2.0).$$

<2> The rare earth magnet according to item <1>, wherein x is $0.15 \leq x \leq 0.30$.

<3> The rare earth magnet according to item <1>, wherein x is $0.10 \leq x \leq 0.25$.

<4> The rare earth magnet according to item <1>, wherein x is $0.15 \leq x \leq 0.25$.

Effects of the Invention

[0014] According to the present invention, in an R-Fe-B-based rare earth magnet, part of Ce of the main phase is replaced by La and the replacement ratio x of Ce by La is specified to a certain range, whereby a rare earth magnet having an enhanced magnetization without using Nd as R can be provided.

Brief Description of the Drawings

[0015]

[Fig. 1] Fig. 1 illustrates a target material used in the sputtering method.

[Fig. 2] Fig. 2 is a view illustrating a targeted composition distribution of a thick film.

[Fig. 3] Fig. 3 is a view illustrating evaluation positions of a thick film.

[Fig. 4] Fig. 4 is a view illustrating the XRD analysis results of the sample where x is from 0.05 to 0.17.

[Fig. 5] Fig. 5 is a view illustrating the relationship between the measurement position and the composition analysis results, regarding the sample where x is from 0.05 to 0.17.

[Fig. 6] Fig. 6 is a view illustrating the relationship between the measurement position and the composition analysis results, regarding the sample where x is from 0.19 to 0.37.

[Fig. 7] Fig. 7 is a view illustrating the MOKE measurement results in a region where x is small.

[Fig. 8] Fig. 8 is a view illustrating the MOKE measurement results in a region where x is medium.

[Fig. 9] Fig. 9 is a view illustrating the relationship between x and the signal strength.

[Fig. 10] Fig. 10 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magne-

tometer in (a) of Fig. 9.

[Fig. 11] Fig. 11 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (b) of Fig. 9.

[Fig. 12] Fig. 12 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (c) of Fig. 9.

[Fig. 13] Fig. 13 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (d) of Fig. 9.

[Fig. 14] Fig. 14 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (e) of Fig. 9.

[Fig. 15] Fig. 15 is a view illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (f) of Fig. 9.

Mode for Carrying Out the Invention

[0016] The embodiment of the rare earth magnet according to the present invention is described in detail below. The present invention is not limited to the following embodiment.

[0017] The R-Fe-B-based rare earth magnet has a main phase and a grain boundary phase surrounding the main phase. The main phase is an $R_2Fe_{14}B$ phase and is a magnetic phase. The grain boundary phase is an R-rich phase and is a non-magnetic phase. Since the $R_2Fe_{14}B$ phase as the magnetic phase is surrounded by the R-rich phase as the non-magnetic phase, each $R_2Fe_{14}B$ phase is magnetically separated, and magnetization inversion is not propagated across a plurality of $R_2Fe_{14}B$ phases. As a result, the rare earth magnet as a whole has excellent magnetization.

[0018] The valence of Ce may be trivalence or tetravalence. When the valence of Ce is trivalence, 4f electrons are localized. When the valence of Ce is tetravalence, 4f electrons are not localized.

[0019] When the valence of Ce is tetravalence, the $Ce_2Fe_{14}B$ phase is stable. However, in the tetravalent Ce, 4f electrons contributing to the enhancement of magnetic properties are not localized, and therefore the magnetization of $Ce_2Fe_{14}B$ phase is lower than the magnetization of $Nd_2Fe_{14}B$ phase.

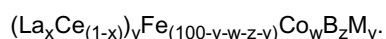
[0020] The present inventors have found that when part of Ce in the $Ce_2Fe_{14}B$ phase is replaced by La and the replacement ratio x of Ce by La is set to a certain range, the magnetization of $(La_xCe_{(1-x)})_2Fe_{14}B$ phase can be significantly enhanced.

[0021] In addition, the present inventors have found that, without being bound by theory, when x in the $(La_xCe_{(1-x)})_2Fe_{14}B$ phase is in a certain range, La acts on Ce to promote localization of 4f electrons in Ce and the valence of Ce is changed from tetravalence to trivalence.

[0022] The configuration of the rare earth magnet of the present invention based on these findings is described below.

(Overall Composition)

[0023] The overall composition of the rare earth magnet of the present invention is represented by the formula:



[0024] In the formula, M is one or more elements selected from Ga, Al, Cu, Au, Ag, Zn, In and Mn, and an unavoidable impurity elements.

[0025] In the formula, x, y, z, w, and v are $0.10 \leq x \leq 0.30$, $5 \leq y \leq 20$, $4 \leq z \leq 6.5$, $0 \leq w \leq 8$, and $0 \leq v \leq 2.0$, respectively. x is the replacement ratio of Ce by La. y is the total content of La and Ce, z is the content of B, w is the content of Co, v is the content of M, and each of y, z, w and v is at%.

[0026] When the formula satisfies $5 \leq y \leq 20$, $4 \leq z \leq 6.5$, $0 \leq w \leq 8$, and $0 \leq v \leq 2.0$, the rare earth magnet becomes a rare earth magnet having a main phase and a grain boundary phase surrounding the main phase. At this time, the main phase is a $(La_xCe_{(1-x)})_2(Fe,Co)_{14}B$ phase, and the grain boundary phase is a (La,Ce)-rich phase. Most of M is present in the grain boundary.

[0027] Respective components in the formula, i.e., La, Ce, Fe, Co, B and M, are described below.

(La)

[0028] La is a rare earth element and, in the rare earth magnet, is present in both the main phase and the grain boundary phase. In the main phase, La acts on Ce, and the valence of Ce is changed from tetravalence to trivalence, whereby the magnetization of the main phase is enhanced.

[0029] In the main phase, La is present in the form of being substituted on Ce. With respect to the replacement ratio x of Ce by La, x in the overall composition is almost the same as x in the main phase.

[0030] When x is 0.1 or more, the effect of changing the valence of Ce from tetravalence to trivalence is clearly developed by La, as a result, magnetization is enhanced (hereinafter, sometimes referred to as "magnetization enhancement effect"). In view of magnetization enhancement effect, x is more preferably 0.15 or more.

[0031] When x is about 0.2, the magnetization enhancement effect reaches a peak. After passing the peak, the magnetization enhancement effect decreases along with an increase in x.

[0032] When x is 0.30 or less, the magnetization enhancement effect is recognized. From the viewpoint that the magnetization enhancement effect is more clearly recognized, x is more preferably 0.25 or less.

[0033] The Curie temperature of the $\text{La}_2\text{Fe}_{14}\text{B}$ is higher than that of the $\text{Ce}_2\text{Fe}_{14}\text{B}$ phase, and therefore it can also be expected that magnetization at high temperatures is enhanced by the replacement of Ce by La.

[0034] In the grain boundary phase, La constitutes a (La,Ce)-rich phase together with Ce. The (La,Ce)-rich phase is a non-magnetic phase and magnetically separates main phases from each other.

(Ce)

[0035] Ce is a rare earth element and in the rare earth magnet, is present in both the main phase and the grain boundary phase. In the main phase, magnetization of the $\text{Ce}_2\text{Fe}_{14}\text{B}$ phase is small, but magnetization of the $(\text{La}_x\text{Ce}_{1-x})_2\text{Fe}_{14}\text{B}$ phase is enhanced when x is in the above-described range.

[0036] In the overall composition, the total content of La and Ce is y at%. When y is from 5 to 20 at%, a main phase in an amount required as a rare earth magnet can be ensured. y may be 7 at% or more, 9 at% or more, or 11 at% or more, and may be 18 at% or less, 16 at% or less, or 14 at% or less.

[0037] In the grain boundary phase, Ce constitutes a (La,Ce)-rich phase together with La. The (La,Ce)-rich phase is a non-magnetic phase and magnetically separates main phases from each other.

(Fe)

[0038] Fe is a main component of the R-Fe-B-based rare earth magnet and constitutes a main phase together with La, Ce and B. In the overall composition, the content of Fe is indicated by the remainder other than La, Ce, Co, B and M.

(Co)

[0039] Co is classified into an iron group element and in the rare earth magnet, Co replaces Fe. In this description, unless otherwise specified, for example, in the $(\text{La}_x\text{Ce}_{1-x})_2\text{Fe}_{14}\text{B}$ phase, even when Co is not indicated, part of Fe in the $(\text{La}_x\text{Ce}_{1-x})_2\text{Fe}_{14}\text{B}$ phase may be replaced by Co. Co is added, if desired, because it improves the heat resistance of the rare earth magnet. In the overall composition, the content w of Co is from 0 to 8 at%. From the viewpoint of improving the heat resistance, the content w of Co may be 1 at% or more, 2 at% or more, or 3 at% or more. In view of saturated enhancement of heat resistance and profitability, the content y of Co may be 7 at% or less, 6 at% or less, or 5 at% or less.

(B)

[0040] B constitutes a main phase together with La, Ce and Fe. When the content z of B in the overall composition is from 4 to 6.5 at%, a main phase necessary for the rare earth magnet can be ensured. z may be 4.2 at% or more, 4.5 at% or more or 4.8 at% or more, and may be 6.2 at% or less, 5.9 at% or less, or 5.6 at% or less.

[0041] M is one or more elements selected from Ga, Al, Cu, Au, Ag, Zn, In, and Mn. M is contained, if desired, to an extent that the effects of the rare earth magnet of the present invention are not impaired. Other than these, M contains an unavoidable impurity. The unavoidable impurity means an impurity that is incapable of avoiding being contained or involves a significant increase in the production cost for avoiding it, such as impurity contained in raw materials. In the overall composition, when the content v of M is from 0 to 2 at%, the effects of the present invention are not impaired. The content v of M may be 0.05 at% or more, 0.1 at% or more, or 0.2 at% or more, and may be 1.0 at% or less, 0.8 at% or less, or 0.5 at% or less.

(Form of Rare Earth Magnet)

[0042] As long as the composition, etc. described above are satisfied, the form of the rare earth magnet is not particularly limited. The form of the rare earth magnet includes a cast, a quenched body, a thin film, a thick film, etc. The quenched body includes a ribbon, a flake, a powder, etc.

(Production Method of Rare Earth Magnet)

[0043] As long as the composition, etc. described above are satisfied, the production method of the rare earth magnet is not particularly limited. The production method includes casting, a liquid quenching method, a sputtering method, etc.

Examples

[0044] The present invention is described more specifically below by referring to Examples. The present invention is not limited to the conditions employed in the following Examples.

(Preparation of Sample)

[0045] A thin film having a composition of $(\text{La}_x\text{Ce}_{1-x})_{11.76}\text{Fe}_{82.36}\text{B}_{5.88}$ was prepared on an SiO_2 substrate by a sputtering method. The film thickness was 500 nm.

[0046] Fig. 1 illustrates a target material used in the sputtering method. The target material 1 has a Ce-Fe-B alloy sheet 2 and an La-Fe-B alloy sheet 3, and the Ce-Fe-B alloy sheet 2 and the La-Fe-B alloy sheet 3 are, as shown in Table 1, joined to each other. The composition of the Ce-Fe-B alloy sheet 2 was $\text{Ce}_{19.5}\text{Fe}_{72.3}\text{B}_{8.2}$, and the composition of the La-Fe-B alloy sheet 3 was $\text{La}_{19.5}\text{Fe}_{72.3}\text{B}_{8.2}$.

[0047] The target material and an SiO_2 substrate were charged into a sputtering apparatus, and a thick film was formed. The thick film formed was heat-treated at 450°C during 90 minutes in a vacuum atmosphere.

[0048] Fig. 2 is a view illustrating a targeted composition distribution of the thick film. As illustrated in Fig. 2, the thick film was formed to become a gradient material in which Ce of the $\text{Ce}_2\text{Fe}_{14}\text{B}$ phase is replaced by La gradually from the upper part toward the lower part. Two sheets of the sample were prepared, and in the sample, the replacement ratio x of Ce by La was from 0.05 to 0.17 in one sheet and from 0.19 to 0.37 in the other sheet. The evaluation method of the replacement ratio x is described later.

(Evaluation of Sample)

[0049] Fig. 3 is a view illustrating evaluation positions of the thick film. The position of (1) in Fig. 3 corresponds to the position of $\text{Ce}_2\text{Fe}_{14}\text{B}$ phase of Fig. 2. The position of (9) in Fig. 3 corresponds to the position of $\text{La}_2\text{Fe}_{14}\text{B}$ phase of Fig. 2.

(Identification of Phase)

[0050] With respect to respective positions of (1), (5) and (9) illustrated in Fig. 3, the phase was identified by performing X-Ray Diffraction (XRD) analysis. The analysis was performed on two sheets of the sample.

[0051] Fig. 4 illustrates the analysis results of the sample where x is from 0.05 to 0.17. In Fig. 4, "2-14-1" means a $(\text{La}_x\text{Ce}_{1-x})_2\text{Fe}_{14}\text{B}$ phase. As shown in Fig. 4, it is confirmed that a $(\text{La}_x\text{Ce}_{1-x})_2\text{Fe}_{14}\text{B}$ phase could be formed other than the position of (1) of Fig. 2, i.e., one end part of the thick film. The same is true for the sample where x is from 0.19 to 0.37.

(Composition Analysis of Phase)

[0052] The composition analysis of the thick film was performed using Energy Dispersive X-ray spectrometry.

[0053] The composition analysis results of the sample where x is from 0.05 to 0.17 are shown in Table 1 and Fig. 5, and the composition analysis results of the sample where x is from 0.19 to 0.37 are shown in Table 2 and Fig. 6.

[Table 1]

	Ce (at%)	La (at%)	Fe (at%)	x
(1)	13.4	0.5	86.0	-
(2)	17.6	1.0	81.4	0.05
(3)	16.6	1.2	82.2	0.07
(4)	15.0	1.6	83.4	0.10
(5)	14.2	2.0	83.8	0.12
(6)	14.0	2.5	83.5	0.15
(7)	13.4	2.7	83.9	0.17

(continued)

	Ce (at%)	La (at%)	Fe (at%)	x
(8)	13.3	2.7	83.9	0.17
(9)	13.1	2.3	84.6	-

[Table 2]

	Ce (at%)	La (at%)	Fe (at%)	x
(1)	11.4	1.9	86.7	-
(2)	13.4	3.2	83.4	0.19
(3)	13.9	3.8	82.3	0.22
(4)	13.7	4.3	82.1	0.24
(5)	13.3	4.6	82.1	0.26
(6)	13.0	5.7	81.3	0.31
(7)	12.3	6.3	81.3	0.34
(8)	11.5	6.6	81.9	0.37
(9)	10.8	6.3	82.9	-

[0054] It can be confirmed from Table 1 and Fig. 5 and from Table 2 and Fig. 6 that both sheets of the sample were a gradient material.

[0055] With respect to respective positions of (1) to (9) of Fig. 3, the magnetic properties were evaluated. As for the evaluation method, an MOKE (Magneto-Optical Kerr Effect) measurement and a measurement using a Vibrating Sample Magnetometer (VSM) were employed.

[0056] The MOKE measurement is a measuring method utilizing a magnetic Kerr effect. The magnetic Kerr effect indicates such an effect that when a surface of a magnetized material is irradiated with linearly polarized light, the intensity of reflected light is changed according to magnetization. In the MOKE measurement, this change in reflected light is evaluated by means of a detector, whereby magnetization of the material is measured. By employing MOKE measurement, the relationship between the composition and magnetic properties of a sample in which the concentration of La has a continuous concentration gradient can be correctly grasped. In the measurement this time, the region of a diameter of 20 μm (the diameter of light with which irradiated) could be measured. In addition, for reference, a measurement using a general vibrating sample magnetometer was also performed.

[0057] Figs. 7 to 15 show the results. Fig. 7 is a view illustrating the MOKE measurement results in a region where x is small. Fig. 8 is a view illustrating the MOKE measurement results in a region where x is medium. Fig. 9 is a view illustrating the relationship between x and the signal strength. Figs. 10 to 15 are views illustrating the measurement results (hysteresis curve) using a vibrating sample magnetometer in (a) to (f) of Fig. 9. Here, from the hysteresis curves of Figs. 10 to 15, the numerical value of magnetization (119.9 emu/g) of Fig. 13 was normalized as a reference value in accordance with the signal strength (0.085) by MOKE measurement of (d) of Fig. 9. Then, respective numerical values of magnetization of Fig. 10, Fig. 11, Fig. 12, Fig. 14 and Fig. 15 were converted by normalization to the signal strength of Fig. 9.

[0058] As shown in Figs. 7 and 8, it could be confirmed that since a Kerr effect is utilized in the MOKE measurement, as the magnetization is higher, the signal strength becomes higher.

[0059] From Fig. 9, with respect to both the MOKE measurement results and the measurement results using a vibrating sample magnetometer, the followings are understood. More specifically, it could be confirmed that the signal starts becoming strong from when x is 0.1, the signal strength reaches a peak when x is around 0.2, the signal strength is thereafter gradually weakened, and the signal strength when x is 0.3 drops to the same level as the signal strength when x is 0.1. It could be also confirmed that the magnetization starts becoming strong from when x is 0.1, the magnetization reaches a peak when x is around 0.2, the magnetization is thereafter gradually weakened, and the magnetization when x is 0.3 drops to the same level as the magnetization when x is 0.1. Furthermore, it could be confirmed that a gradual change in the composition of the thick film can be more correctly measured in the MOKE measurement than in the measurement using a vibrating sample magnetometer.

[0060] From these results, the effects of the present invention could be verified.

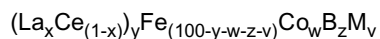
Description of Numerical References

[0061]

- 5 1 Target material
 2 Ce-Fe-B Alloy sheet
 3 La-Fe-B Alloy sheet

10 **Claims**

1. A rare earth magnet having an overall composition represented by the formula:



- 15 (wherein M is one or more elements selected from Ga, Al, Cu, Au, Ag, Zn, In and Mn, and an unavoidable impurity elements, and
 x, y, z, w and v satisfy the following relationship:

$$20 \qquad 0.10 \leq x \leq 0.30,$$

$$25 \qquad 5 \leq y \leq 20,$$

$$4 \leq z \leq 6.5,$$

$$30 \qquad 0 \leq w \leq 8,$$

and

$$35 \qquad 0 \leq v \leq 2.0).$$

2. The rare earth magnet according to claim 1, wherein x is $0.15 \leq x \leq 0.30$.

- 40 3. The rare earth magnet according to claim 1, wherein x is $0.10 \leq x \leq 0.25$.

4. The rare earth magnet according to claim 1, wherein x is $0.15 \leq x \leq 0.25$.

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FIG. 1

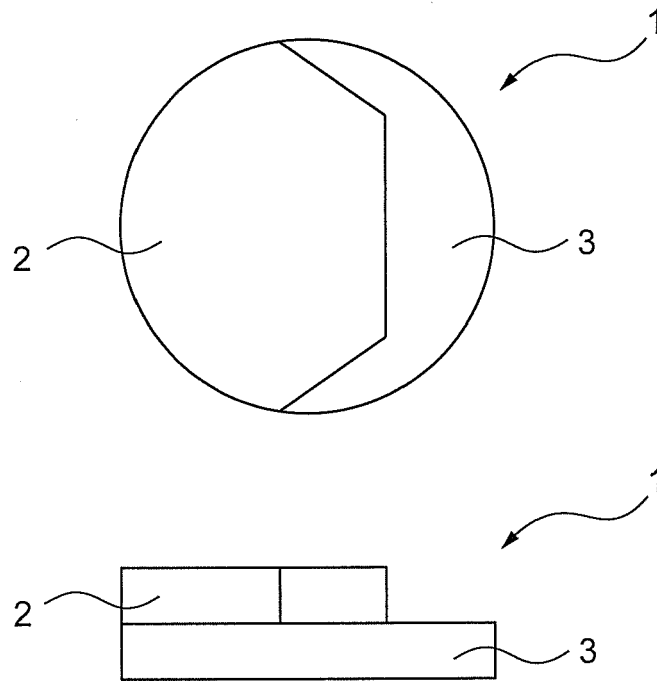


FIG. 2

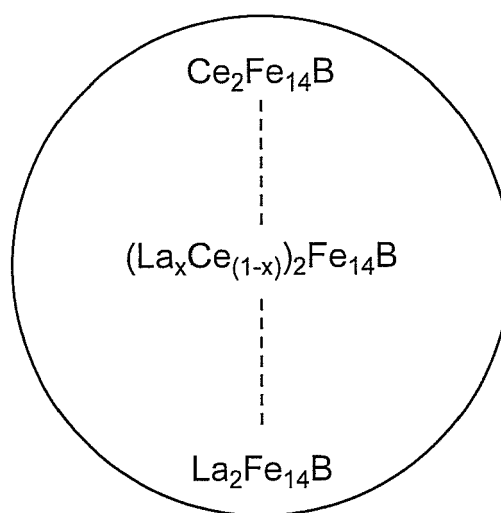


FIG. 3

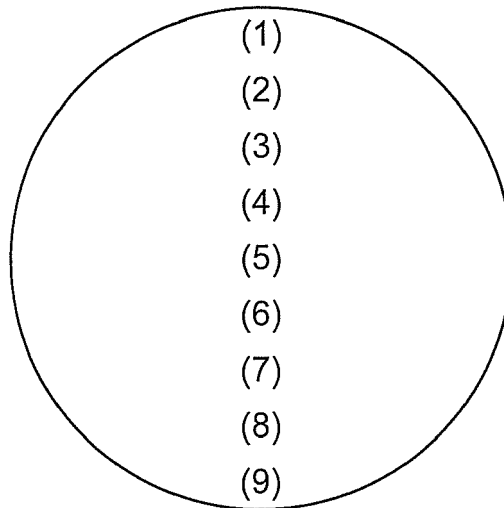


FIG. 4

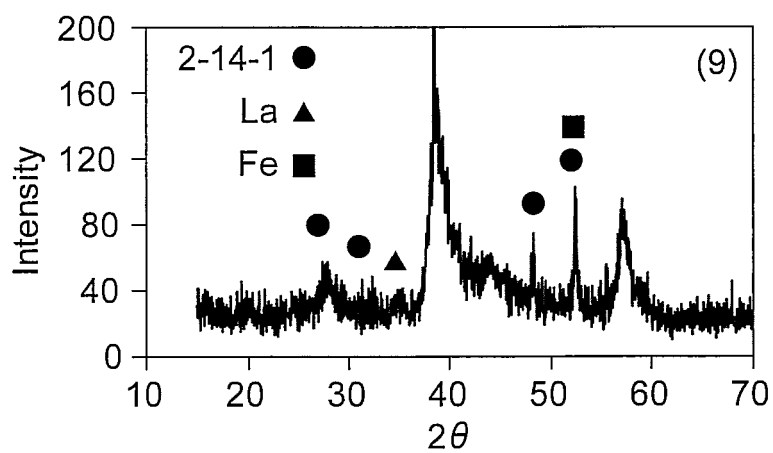
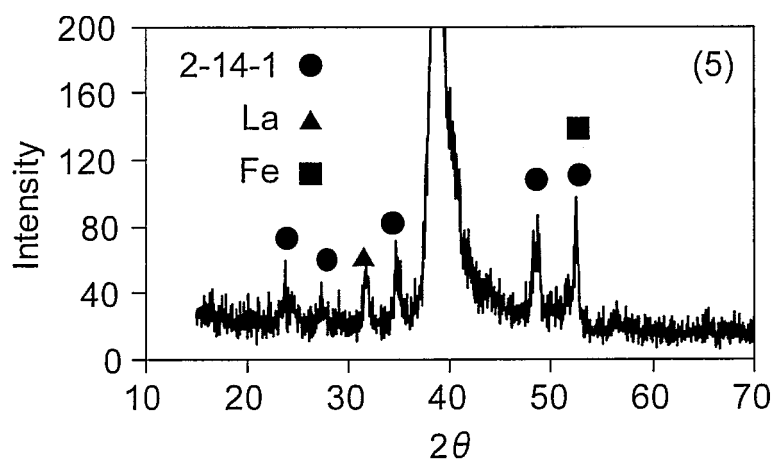
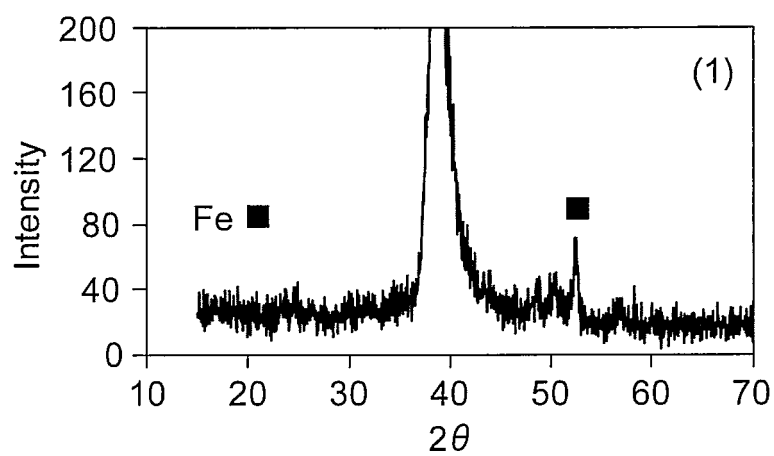


FIG. 5

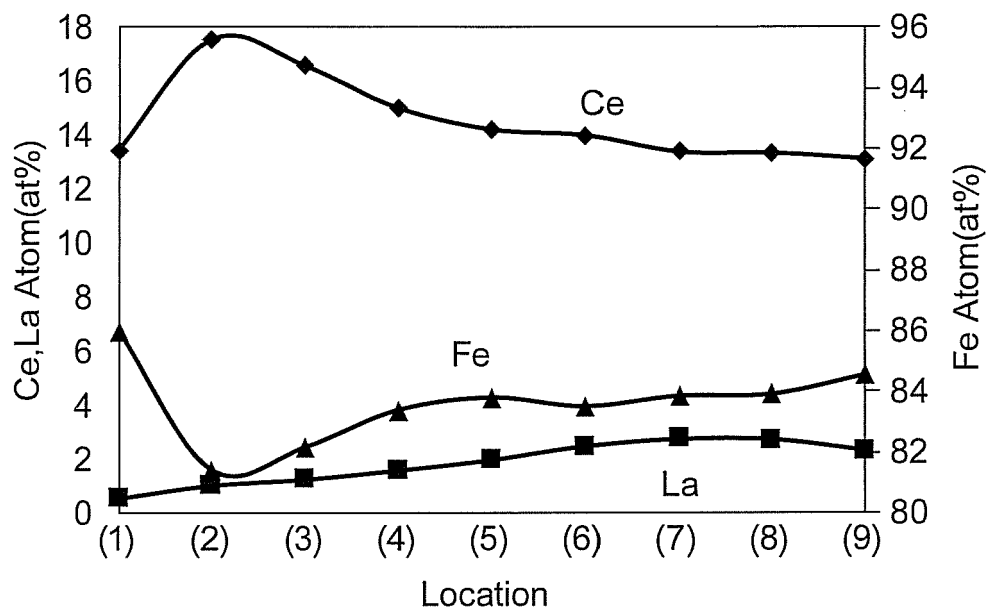


FIG. 6

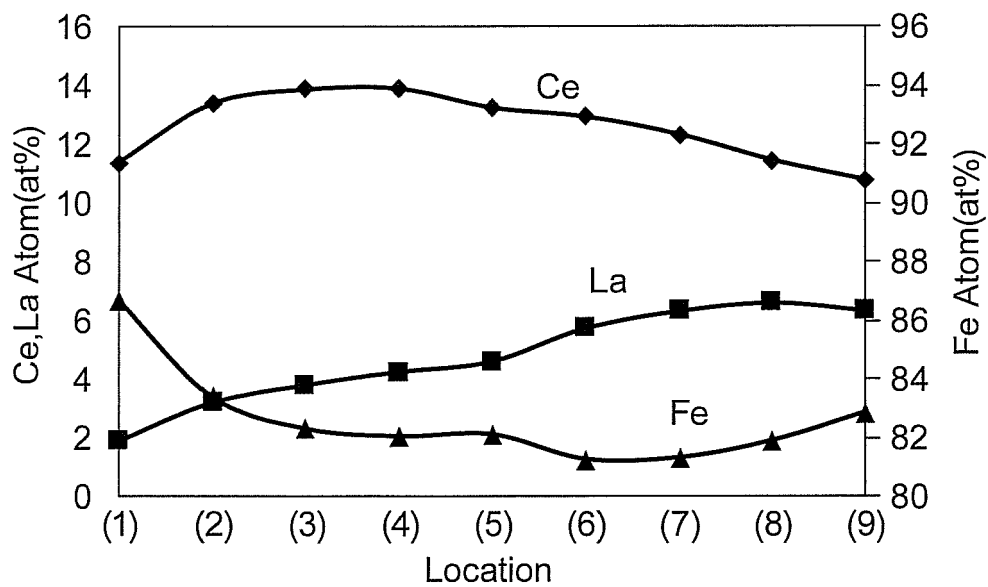


FIG. 7

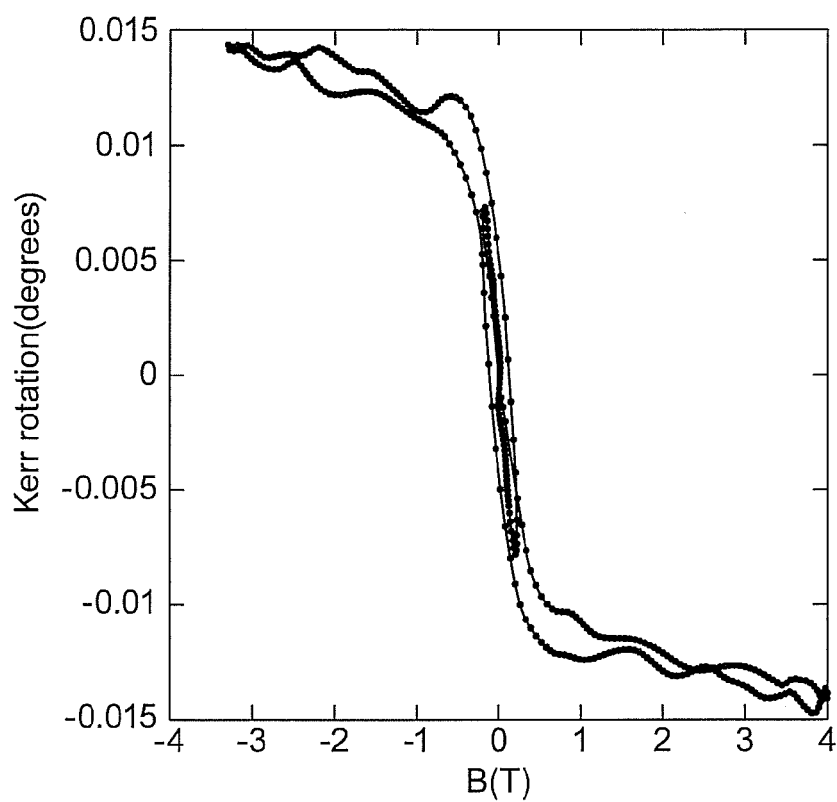


FIG. 8

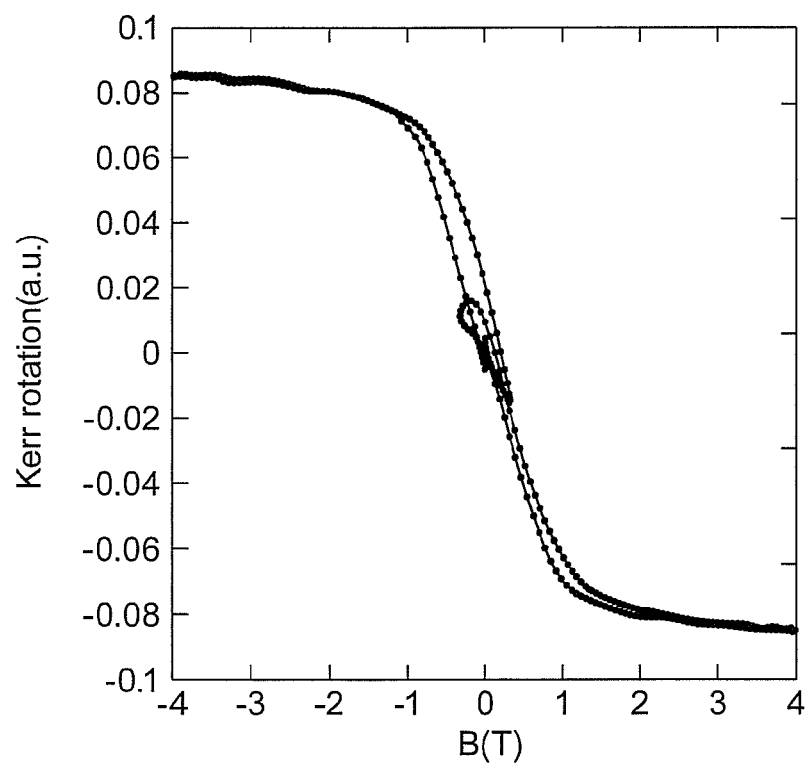


FIG. 9

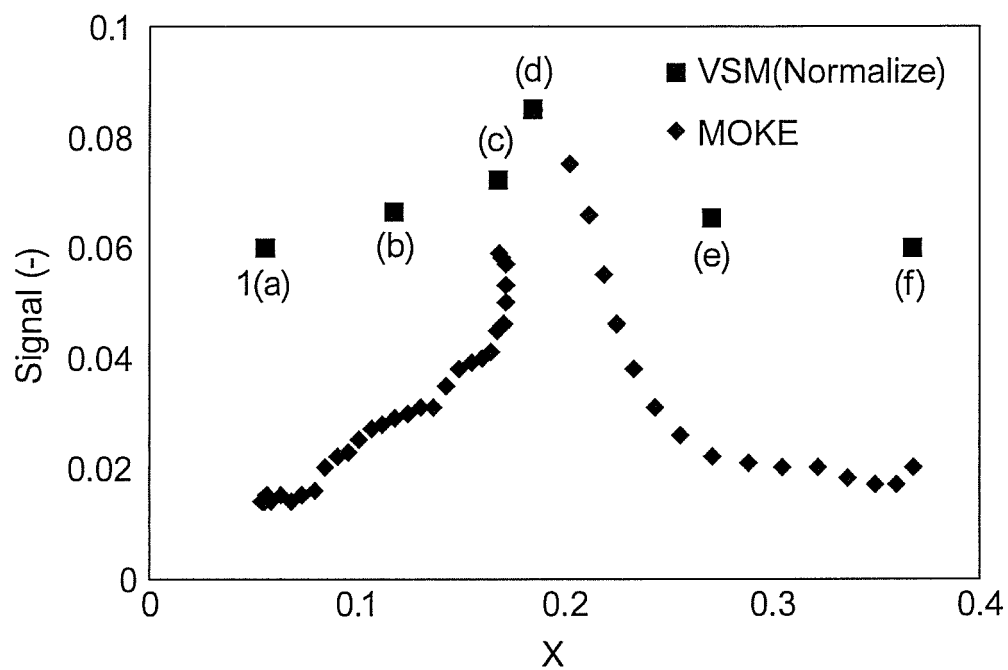


FIG. 10

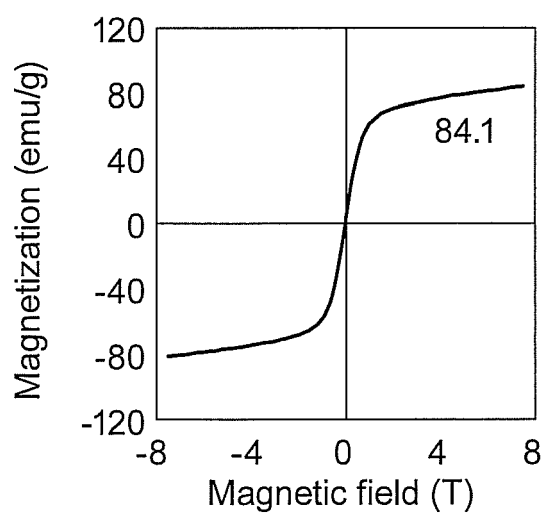


FIG. 11

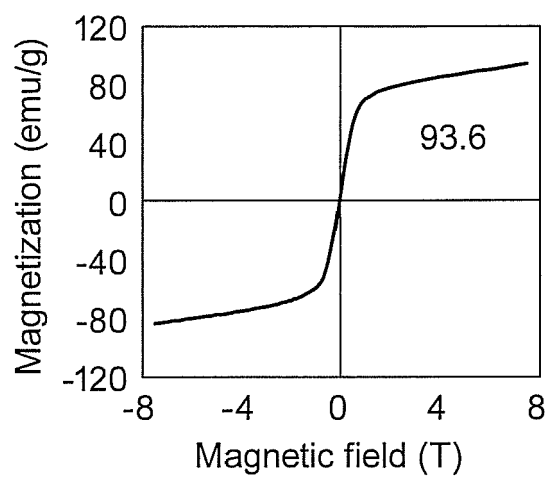


FIG. 12

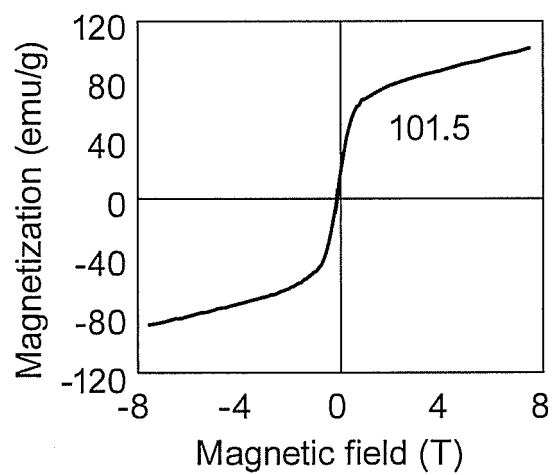


FIG. 13

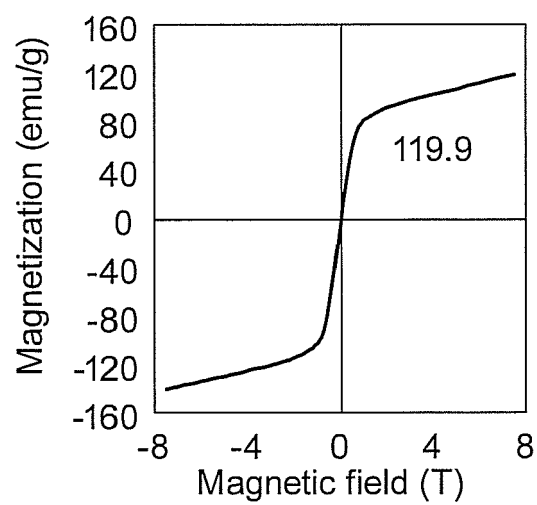


FIG. 14

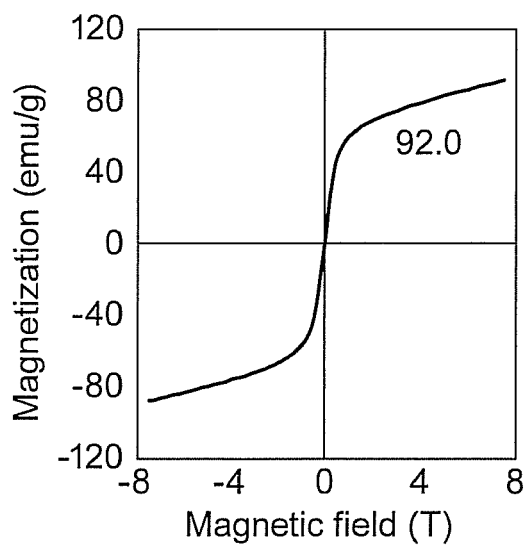
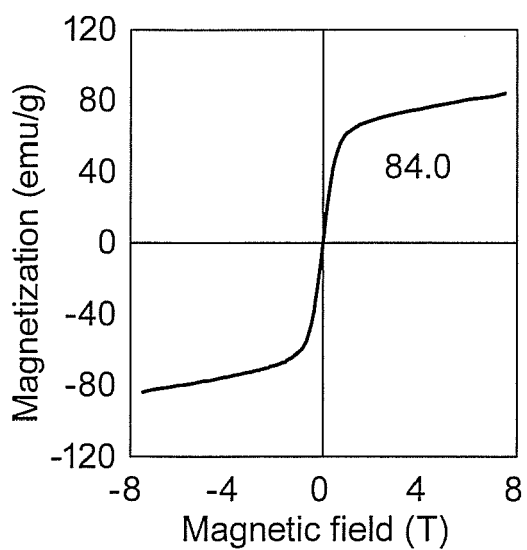


FIG. 15





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Place of search Munich		Date of completion of the search 19 May 2017	Examiner Primus, Jean-Louis
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