

DOUBLE-SHELL NEODYMIUM-IRON-BORON MAGNET AND PREPARATION METHOD THEREFOR

FIELD OF THE INVENTION

5 The invention relates to a double-shell neodymium-iron-boron magnet and the preparation method thereof.

BACKGROUND OF THE INVENTION

10 In recent years, with the continuous improvement of green travel, energy saving and environmental protection, the demand for the sintered NdFeB magnets with high coercivity has been increasing for electric vehicles, compressors of inverter air conditioners and wind power generation. At present, the preparation of the sintered NdFeB magnets with high coercivity is mainly achieved by replacing light rare earth elements with heavy rare earth elements Dy and/or Tb. On the one hand, this increases the cost of raw materials for
15 sintering NdFeB. In addition, the addition of heavy rare earth elements further reduces the remanence of the magnet and sacrifices a part of the magnetic energy product of the magnet.

 In the prior art, the performance of magnets can be improved by a double alloy method. The double alloy method comprises smelting the main alloy and the auxiliary alloy separately, mixing and sintering the materials after crushing, and modifying the grain
20 boundaries of the alloy particles in the main phase by changing the composition and ratio of the auxiliary alloy powder. However, due to the higher temperature in the sintering stage, the heavy rare earth elements such as Dy and Tb added as auxiliary phases will diffuse into the main phase in large quantities, resulting in a decrease in the remanence of the magnet. At the same time, a large amount of heavy rare earth elements diffuse into the main phase, and the
25 concentration of heavy rare earth elements in the outer shell of the main phase particles decreases, resulting in that the improvement value for the coercive force by heavy rare earth elements is less than the effect of heavy rare earth elements distributed on the surface of grains to improve the structure of grain boundaries, which will lead to low utilization of heavy rare earths and limited improvement for coercivity.

30 Patent document CN111636035A discloses a heavy rare earth alloy, a

neodymium-iron-boron permanent magnet material, a raw material and a preparation method. The auxiliary alloy components are heavy rare earth metals, B, Ti and/or Zr, Fe and/or Co and the like. By controlling the contents of Ti and/or Zr and the total amount of heavy rare earth elements or the like, Ti and/or Zr are combined with B, so as to avoid excessive heavy rare earth metals to be combined with B, and reduce the amount of heavy rare earth metals diffused into the main phase, so that the performance of the magnet is improved. But this solution needs further optimization. The amount of heavy rare earths such as Dy and Tb diffused into the main phase is still quite large, and the thickness of the shell formed by these heavy rare earth elements on the outer edge of the main phase is relatively deep.

Therefore, it is necessary to find a new process, which can effectively make more expensive heavy rare earth elements such as Dy and Tb to form a thinner shell around the main phase, so as to reduce the diffusion degree of heavy rare earth elements into the main phase.

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SUMMARY OF THE INVENTION

In order to solve the problem that a large amount of heavy rare earth elements diffuse into the main phase in the double alloy method in the prior art, the present invention provides double-shell neodymium-iron-boron magnet and the preparation method thereof. The invention can achieve a double-shell neodymium-iron-boron magnet, wherein a thinner shell is formed around the main phase, which effectively reduces the diffusion amount of heavy rare earth elements into the main phase.

The present invention solves the above-mentioned technical problem through the following technical solutions.

The invention provides a double-shell neodymium-iron-boron magnet, comprising a main phase crystal grain, a double shell thereof and an Nd-rich phase adjacent to the main phase crystal grain,

the main phase crystal grain comprises $R_2Fe_{14}B$, wherein the R is one or more of La, Ce, Pr and Nd;

the double shell has an inner layer comprising $(Nd/Ho)_2Fe_{14}B$ and/or $(Nd/Gd)_2Fe_{14}B$;

the double shell has an outer layer comprising $(\text{Nd/Dy})_2\text{Fe}_{14}\text{B}$ and/or $(\text{Nd/Tb})_2\text{Fe}_{14}\text{B}$;
the double shell has a thickness of 0.1-6 μm ;

the Nd-rich phase comprises a $(\text{R-RH})_6\text{T}_{13}\text{X}$ phase, wherein RH is one or more of Ho, Gd, Dy and Tb; T is Fe and/or Co; and X is one or more of Ga, Cu and Al.

5 In the invention, preferably, the double shell has an inner layer comprising $(\text{Nd/Ho})_2\text{Fe}_{14}\text{B}$.

In the invention, preferably, the double shell has an outer layer comprising $(\text{Nd/Dy})_2\text{Fe}_{14}\text{B}$.

In the invention, preferably, the Nd-rich phase further comprises ZrB_2 and TiB_2 .

10 The invention further provides a preparation method of the double-shell neodymium-iron-boron magnet as mentioned above, comprising the steps of:

S1: preparing a main alloy sheet, a first auxiliary alloy sheet and a second auxiliary alloy sheet respectively;

Wherein, the raw material for the first auxiliary alloy sheet comprises LH_1 , RH_1 , X_1 and Fe, wherein the LH_1 is one or more of La, Ce, Pr and Nd; the RH_1 is Ho and/or Gd; the X_1 is one or more of Cu, Co, Ga and Al; in the first auxiliary alloy sheet, the LH_1 accounts for 0-80% by mass of the first auxiliary alloy sheet, the RH_1 accounts for 5-80% by mass of the first auxiliary alloy sheet, the total amount of LH_1 and RH_1 accounts for 30% by mass or more of the first auxiliary alloy sheet, and the X_1 accounts for 1-15% by mass of the first auxiliary alloy sheet, wherein the sum of the mass percentages of respective elements in the first auxiliary alloy sheet is 100%;

20 The raw material for the second auxiliary alloy sheet comprises RH_2 , X_2 and Fe, wherein the RH_2 is Dy and/or Tb, the X_2 is Zr and/or Ti; in the second auxiliary alloy sheet, the RH_2 accounts for 0-80% by mass, excluding 0% by mass of the second auxiliary alloy sheet, and the X_2 accounts for 3-10% by mass of the second auxiliary alloy sheet, wherein the sum of the mass percentages of respective elements in the second auxiliary alloy sheet is 100%;

25 S2: subjecting a mixture, which is obtained by hydrogen decrepitating or pulverizing the main alloy sheet, the first auxiliary alloy sheet and the second auxiliary alloy sheet, to a shaping and sintering treatments to achieve the double-shell neodymium-iron-boron magnet.

In S1, preferably, the first auxiliary alloy sheet has a melting point which is lower than that of the second auxiliary alloy sheet.

In S1, preferably, the LH_1 accounts for 0-60% by mass, excluding 0% by mass, for example 30% by mass of the first auxiliary alloy sheet.

5 In S1, preferably, the LH_1 is Pr and/or Nd.

In S1, preferably, when the raw material for the first auxiliary alloy sheet comprises Pr, the Pr accounts for 0-60% by mass, for example, 7.5% by mass of the first auxiliary alloy sheet.

10 In S1, preferably, when the raw material for the first auxiliary alloy sheet comprises Nd, the Nd accounts for 0-60% by mass, for example, 22.5% by mass of the first auxiliary alloy sheet.

In S1, preferably, the RH_1 accounts for 20-50% by mass of the first auxiliary alloy sheet.

15 In S1, preferably, the total amount of LH_1 and RH_1 accounts for 50% by mass or more of the first auxiliary alloy sheet.

In S1, preferably, the X_1 is Cu and/or Co.

In S1, preferably, the X_1 accounts for 5-12% by mass, preferably 5-10% by mass of the first auxiliary alloy sheet.

20 In S1, preferably, when the X_1 comprises Cu, the Cu accounts for 1-6% by mass, for example, 5% by mass of the first auxiliary alloy sheet.

In S1, preferably, when the X_1 comprises Co, the Co accounts for 1-6% by mass, for example, 5% by mass of the first auxiliary alloy sheet.

25 In S1, preferably, in the first auxiliary alloy sheet, the Fe accounts for 50% by mass or less, preferably 39-45% by mass, for example, 39.5% by mass or 44.7% by mass of the first auxiliary alloy sheet.

In S1, preferably, the raw material for the first auxiliary alloy sheet further comprises B, and in the first auxiliary alloy sheet, the B accounts for 0-0.6% by mass, excluding 0% by mass, for example 0.3% by mass or 0.5% by mass of the first auxiliary alloy sheet.

30 In S1, in an preferable embodiment, the raw material for the first auxiliary alloy is composed of the following components: Ho with a content of 50%; Cu with a content of 5%;

Fe with a content of 44.7%; and B with a content of 0.3%, wherein the percentages refer to the mass percentages of the components in the raw material for the first auxiliary alloy.

In S1, in an preferable embodiment, the raw material for the first auxiliary alloy is composed of the following components: Pr with a content of 7.5%; Nd with a content of 22.5%; Ho with a content of 20%; Cu with a content of 5%; Co with a content of 5%; Fe with a content of 39.5%; and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the components in the raw material for the first auxiliary alloy.

In S1, preferably, the first auxiliary alloy sheet is obtained by smelting and casting the raw material for the first auxiliary alloy sheet.

Wherein, preferably, the temperature for smelting the raw material for the first auxiliary alloy sheet is 1000°C or more.

Wherein, the casting for the first auxiliary alloy sheet can be a traditional casting process in the field, for example, a strip continuous casting process, an ingot casting process, a centrifugal casting process, or a rapid quenching process.

In S1, preferably, the RH_2 accounts for 50-80% by mass, for example, 58% by mass or 63% by mass of the second auxiliary alloy sheet.

In S1, preferably, the X_2 accounts for 6-10% by mass of the second auxiliary alloy sheet.

In S1, preferably, when the X_2 comprises Zr, the Zr accounts for 6-10% by mass of the second auxiliary alloy sheet.

In S1, preferably, the raw material for the second auxiliary alloy sheet further comprises LH_2 , the LH_2 is one or more of La, Ce, Pr, and Nd, and the LH_2 accounts for 80% by mass or less, excluding 0% by mass of the second auxiliary alloy sheet.

In S1, preferably, the raw material for the second auxiliary alloy sheet further comprises B, and in the second auxiliary alloy sheet, the B accounts for 0-0.6% by mass, excluding 0% by mass, for example, 0.5% by mass of the second auxiliary alloy sheet.

In S1, in a preferable embodiment, the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 58%; Zr with a content of 6%; and Fe with a content of 36%, wherein the percentages refer to the mass percentages of the components in the raw material for the second auxiliary alloy.

In S1, in a preferable embodiment, the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 63%; Zr with a content of 6%; Fe with a content of 30.5%; and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the components in the raw material for the second auxiliary alloy.

5 In S1, in a preferable embodiment, the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 58%; Zr with a content of 6%; Fe with a content of 35.5%; and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the components in the raw material for the second auxiliary alloy.

In S1, preferably, the second auxiliary alloy sheet is obtained by smelting and casting
10 the raw material for the second auxiliary alloy sheet.

Wherein, preferably, the temperature for smelting the raw material for the second auxiliary alloy sheet is 1300°C or more.

Wherein, the casting for the second auxiliary alloy sheet can be a traditional casting process in the field, for example, a strip continuous casting process, an ingot casting process,
15 a centrifugal casting process, or a rapid quenching process.

In S1, preferably, the raw material for the main alloy sheet comprises LH_3 , X_3 , Y_3 , Fe and B, wherein the LH_3 is Pr and/or Nd; the X_3 is one or more of Zr, Ti and Nb; the Y_3 is one or more of Cu, Al, Ga and Co, and the Y_3 must comprise Cu; and in the main alloy sheet, the LH_3 accounts for 27-35% by mass of the main alloy sheet, the X_3 accounts for 0.05-0.8%
20 by mass of the main alloy sheet, wherein the sum of the mass percentages of respective elements in the main alloy sheet is 100%.

Wherein, preferably, the LH_3 accounts for 27-30% by mass, for example, 27.6% by mass, 28% by mass or 29% by mass of the main alloy sheet.

Wherein, preferably, the X_3 accounts for 0.1-0.6% by mass, preferably 0.2-0.3% by
25 mass, for example 0.22% by mass of the main alloy sheet.

Wherein, preferably, the Y_3 accounts for 0.75-2.5% by mass, for example 1.15% by mass or 2.06% by mass of the main alloy sheet.

Wherein, preferably, in the main alloy sheet, the Cu accounts for 0.1-0.6% by mass, for example 0.21% by mass or 0.25% by mass of the main alloy sheet.

30 Wherein, preferably, in the main alloy sheet, when the Y_3 comprises Al, the Al

accounts for 0.02-1.2% by mass, for example 0.4% by mass of the main alloy sheet.

Wherein, preferably, in the main alloy sheet, when the Y_3 comprises Ga, the Ga accounts for 0.25-0.4% by mass, for example 0.26% by mass of the main alloy sheet.

Wherein, preferably, in the main alloy sheet, when the Y_3 comprises Co, the Co
5 accounts for 0.5-1.2% by mass, for example 1.19% by mass of the main alloy sheet.

Wherein, preferably, in the main alloy sheet, the B accounts for 0.97-1% by mass of the main alloy sheet.

Wherein, preferably, the raw material for the main alloy sheet further comprises RH_3 , the RH_3 is Dy and/or Tb, and the RH_3 accounts for 0-3% by mass, excluding 0% by mass of
10 the main alloy sheet.

In S1, in a preferable embodiment, the raw material for the main alloy is composed of the following components: Nd with a content of 27.6%; Dy with a content of 3%; Ga with a content of 0.26%; Al with a content of 0.4%; Cu with a content of 0.21%; Co with a content of 1.19%; Ti with a content of 0.2%; Nb with a content of 0.02%; B with a content
15 of 1%; Fe with a content of 66.12%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy.

In S1, in a preferable embodiment, the raw material for the main alloy is composed of the following components: Pr with a content of 7%; Nd with a content of 21%; Dy with a content of 3%; Ga with a content of 0.4%; Cu with a content of 0.25%; Co with a content of
20 0.5%; Zr with a content of 0.2%; B with a content of 0.97%; and Fe with a content of 66.68%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy.

In S1, in a preferable embodiment, the raw material for the main alloy is composed of the following components: Pr with a content of 7.25%; Nd with a content of 21.75%; Dy
25 with a content of 1%; Ga with a content of 0.4%; Cu with a content of 0.25%; Co with a content of 0.5%; Zr with a content of 0.2%; B with a content of 0.97%; Fe with a content of 67.68%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy.

In S1, preferably, the main alloy sheet is obtained by smelting and casting the raw
30 material for the main alloy sheet.

Wherein, preferably, the temperature for smelting the raw material for the main alloy sheet is 1400°C or more.

Wherein, the casting for the main alloy sheet can be a traditional casting process in the field, for example, a strip continuous casting process, an ingot casting process, a centrifugal casting process, or a rapid quenching process.

In S2, the amount of the first auxiliary alloy sheet and the second auxiliary alloy sheet accounts for preferably more than 1% by mass and less than 15% by mass, preferably 5% by mass of the amount of the raw material for the double-shell neodymium-iron-boron magnet; the raw material for the double-shell neodymium-iron-boron magnet comprises the main alloy sheet, the first auxiliary alloy sheet and the second auxiliary alloy sheet

In S2, preferably, a mixture of the main alloy sheet and the auxiliary alloy sheets is subjected to hydrogen decrepitation, pulverization, shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet; or

the main alloy sheet and the auxiliary alloy sheets are subjected to hydrogen decrepitation respectively, then the coarse powders obtained after hydrogen decrepitating the main alloy sheet and the auxiliary alloy sheets are mixed, and then the mixed coarse powders are subjected to pulverization, shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet; or

the main alloy sheet and the auxiliary alloy sheets are subjected to hydrogen decrepitation and pulverization respectively, then the coarse powders obtained after pulverizing the main alloy sheet and the auxiliary alloy sheets are mixed, and then the mixed coarse powders are subjected to shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet.

Wherein, the operation and conditions for the hydrogen decrepitation can be conventional in the art. The hydrogen decrepitation has a dehydrogenation temperature of 400°C-650°C, for example, 500-620°C.

Wherein, the process of the pulverization can be a conventional pulverization process in the art, such as, jet mill pulverization, and preferably, the pulverization is carried out under an atmosphere with an oxygen content of 50 ppm or less. The particles after the pulverization have a diameter of 2-7 μm, for example, 3.0-5.3 μm.

Wherein, the conditions for shaping can be conventional in the art, for example, the shaping comprises pressing into a green compact in a press with a magnetic field strength of 0.5T-3.0T, for example, 1.0-2.0T.

Wherein, the conditions of the sintering treatment can be conventional in the field.
5 The sintering temperature can be 1000-1150°C, for example, 1050-1085°C.

On the basis of conforming to common knowledge in the field, the above-mentioned preferred conditions can be combined arbitrarily to obtain the preferred examples of the present invention.

The reagents and raw materials used in the present invention are all commercially
10 available.

The positive progress effects of the present invention are as follows:

The invention achieves a double-shell neodymium-iron-boron magnet, wherein a cheaper heavy rare earth shell of Ho and/or Gd is firstly formed around the main phase, which inhibits the excessive diffusion of Dy and/or Tb expensive rare earth elements into
15 the main phase, allowing Dy and/or Tb to form a thinner shell layer around the main phase, which effectively reduces the diffusion amount of heavy rare earth elements Dy and/or Tb into the main phase, and achieves the effect of reducing the usage amount of Dy and Tb while achieving the same performance.

20 **BRIEF DESCRIPTION OF THE DRAWINGS**

Fig. 1 shows an EMPA diagram of the sample in Example 1.

Fig. 2 shows the composition line scanning result of Ho and Dy elements in the sample in Example 1.

25 **DETAILED DESCRIPTION OF THE INVENTION**

The present invention is further illustrated below by means of examples, but the present invention is not limited to the scope of the examples.

Example 1

30 (1) According to the formulation shown in Table 1, a main alloy sheet, a first

auxiliary alloy sheet and a second auxiliary alloy sheet were prepared, respectively.

Wherein, the temperature for smelting the main alloy sheet was 1400-1600°C, and then the main alloy sheet was obtained through casting by a rapid quenching process.

The temperature for smelting the first auxiliary alloy sheet was 1400-1600°C, and
5 then the first auxiliary alloy sheet was obtained through casting by a rapid quenching process.

The temperature for smelting the second auxiliary alloy sheet was 1400-1600°C, and then the second auxiliary alloy sheet was obtained through casting by a rapid quenching process.

10 (2) Process of Hydrogen Decrepitation: A mixture of the main alloy sheet, the first auxiliary alloy sheet and the second auxiliary alloy sheet prepared in Step (1) at room temperature was subjected to hydrogen decrepitation treatment at 500-620°C to obtain a coarsely pulverized powder.

(3) Pulverization Treatment: The coarsely pulverized powder from Step (2) was
15 finely pulverized in an air jet mill in an atmosphere with an oxygen content of 50 ppm or less to obtain a finely pulverized powder with an average particle diameter of $D_{50} = 3.0-5.3 \mu\text{m}$.

(4) Shaping Process: The finely pulverized powder was pressed in a press with a magnetic field strength of 1.0-2.0T to form a green compact, which was then kept under the
20 condition with a pressure of 260 MPa for 15s to obtain a molded body.

(5) Sintering Process: The molded body was sintered at a temperature of 1050-1085°C in a sintering atmosphere being vacuum or argon atmosphere to obtain a double-shell neodymium-iron-boron magnet.

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Table 1 The amounts of respective elements in the main alloy sheet, the first auxiliary alloy sheet and the second auxiliary alloy sheet in Examples 1-3.

	Elements	Example 1	Example 2	Example 3
Main Alloy Sheet (Sheet A)	Pr	/	7	7.25
	Nd	27.6	21	21.75
	Dy	3	3	1
	Ga	0.26	0.4	0.4
	Al	0.4	/	/
	Cu	0.21	0.25	0.25
	Co	1.19	0.5	0.5
	Ti	0.2	/	/
	Zr	/	0.2	0.2
	Nb	0.02	/	/
	B	1	0.97	0.97
	Fe	66.12	66.68	67.68
First Auxiliary Alloy Sheet (Sheet B)	Pr	/	7.5	7.5
	Nd	/	22.5	22.5
	Ho	50	20	20
	Cu	5	5	5
	Co	/	5	5
	Fe	44.7	39.5	39.5
	B	0.3	0.5	0.5
Second Auxiliary Alloy Sheet (Sheet C)	Dy	58	63	58
	Zr	6	6	6
	Fe	36	30.5	35.5
	B	/	0.5	0.5
Mass Ratio of Sheet A : Sheet B : Sheet C		95:2:3	95:3:2	95:2:3

5 Wherein, “/” means that this component is not comprised.

Examples 2-3

Except for the different amounts of raw materials shown in Table 1, other process conditions were the same as in Example 1 to obtain the double-shell neodymium-iron-boron magnets.

Effect Example

Magnetic performance test: The magnetic properties of the double-shell neodymium-iron-boron magnets were tested by using the PFM14.CN molding type ultra-high coercivity permanent magnet measuring instrument of China Metrology Institute.

	Example 1	Example 2	Example 3
Whether shell structure is formed	Yes	Yes	Yes
Thickness of Shell (μm)	0.1~6	0.1~6	0.1~6
Coercivity (kOe)	29.3	28.7	24.0
Remanence (kGs)	12.3	12.6	13.2

In the present invention, by adjusting the formulations of the first auxiliary alloy sheet, the second auxiliary alloy sheet and the main alloy sheet so that the melting point of the first auxiliary alloy sheet is lower than the melting point of the second auxiliary alloy sheet, thus in the sintered process, the first auxiliary alloy sheet is melted earlier than the second auxiliary alloy sheet to diffuse around the main phase and forms an inner layer of the double-shell. Then, an outer layer of the double shell with Dy and/or Tb as the heavy rare earth elements is further formed so that Dy and/or Tb can only form a shell layer at the outermost periphery.

FIG. 1 is the result of the EPMA diagram of the sample of Example 1. Dy forms a clear shell layer with a high content in the outer layer of the main phase, which can achieve the effect of increasing H_{cj} . From the composition line scanning results of Example 1 in Fig. 2, it can be seen that the diffusion degree of Ho is close to that of Dy. This phenomenon shows that both Ho and Dy will preferentially replace Nd at the 4f position in the main

phase. In the present invention, the addition of the Ho element to occupy a part of the diffusion channel of the heavy rare earth elements reduces the diffusion amount of the heavy rare earth element Dy from the outer layer of the main phase to the inner layer, thereby achieving the effect of increasing the Dy concentration in the outer layer of the main phase.

WHAT IS CLAIMED IS:

1. A preparation method of a double-shell neodymium-iron-boron magnet, comprising the steps of:

S1: preparing a main alloy sheet, a first auxiliary alloy sheet and a second auxiliary alloy sheet respectively;

wherein, the raw material for the main alloy sheet comprises LH_3 , X_3 , Y_3 , Fe and B, wherein the LH_3 is Nd, or Pr and Nd; the X_3 is one or more of Zr, Ti and Nb; the Y_3 is one or more of Cu, Al, Ga and Co, and the Y_3 must comprise Cu; and in the main alloy sheet, the LH_3 accounts for 27-35% by mass of the main alloy sheet, the X_3 accounts for 0.05-0.8% by mass of the main alloy sheet, wherein the sum of the mass percentages of respective elements in the main alloy sheet is 100%,

the raw material for the first auxiliary alloy sheet comprises LH_1 , RH_1 , X_1 and Fe, wherein the LH_1 is one or more of La, Ce, Pr and Nd; the RH_1 is Ho and/or Gd; the X_1 is one or more of Cu, Co, Ga and Al; in the first auxiliary alloy sheet, the LH_1 accounts for 0-80% by mass of the first auxiliary alloy sheet, the RH_1 accounts for 5-80% by mass of the first auxiliary alloy sheet, the total amount of LH_1 and RH_1 accounts for 30% by mass or more of the first auxiliary alloy sheet, and the X_1 accounts for 1-15% by mass of the first auxiliary alloy sheet, wherein the sum of the mass percentages of respective elements in the first auxiliary alloy sheet is 100%;

the raw material for the second auxiliary alloy sheet comprises RH_2 , X_2 and Fe, wherein the RH_2 is Dy and/or Tb, the X_2 is Zr and/or Ti; in the second auxiliary alloy sheet, the RH_2 accounts for 0-80% by mass, excluding 0% by mass of the second auxiliary alloy sheet, and the X_2 accounts for 3-10% by mass of the second auxiliary alloy sheet, wherein the sum of the mass percentages of respective elements in the second auxiliary alloy sheet is 100%;

S2: subjecting a mixture, which is obtained by hydrogen decrepitating or pulverizing the main alloy sheet, the first auxiliary alloy sheet and the second auxiliary alloy sheet, to a shaping and sintering treatments to achieve the double-shell neodymium-iron-boron magnet.

2. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 1, wherein:

in S1, the LH_1 accounts for 0-60% by mass, excluding 0% by mass, for example 30% by mass of the first auxiliary alloy sheet; and/or

5 in S1, the LH_1 is Pr and/or Nd; and/or

in S1, when the raw material for the first auxiliary alloy sheet comprises Pr, the Pr accounts for 0-60% by mass, for example, 7.5% by mass of the first auxiliary alloy sheet; and/or

10 in S1, when the raw material for the first auxiliary alloy sheet comprises Nd, the Nd accounts for 0-60% by mass, for example, 22.5% by mass of the first auxiliary alloy sheet; and/or

in S1, the RH_1 accounts for 20-50% by mass of the first auxiliary alloy sheet; and/or

in S1, the total amount of LH_1 and RH_1 accounts for 50% by mass or more of the first auxiliary alloy sheet; and/or

15 in S1, the X_1 is Cu and/or Co; and/or

in S1, the X_1 accounts for 5-12% by mass, preferably 5-10% by mass of the first auxiliary alloy sheet; and/or

in S1, when the X_1 comprises Cu, the Cu accounts for 1-6% by mass, for example, 5% by mass of the first auxiliary alloy sheet; and/or

20 in S1, when the X_1 comprises Co, the Co accounts for 1-6% by mass, for example, 5% by mass of the first auxiliary alloy sheet; and/or

in S1, in the first auxiliary alloy sheet, the Fe accounts for 50% by mass or less, preferably 39-45% by mass, for example, 39.5% by mass or 44.7% by mass of the first auxiliary alloy sheet; and/or

25 in S1, the raw material for the first auxiliary alloy sheet further comprises B, and in the first auxiliary alloy sheet, the B accounts for 0-0.6% by mass, excluding 0% by mass, for example 0.3% by mass or 0.5% by mass of the first auxiliary alloy sheet.

3. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 2, wherein:

the raw material for the first auxiliary alloy is composed of the following components: Ho with a content of 50%; Cu with a content of 5%; Fe with a content of 44.7%; and B with a content of 0.3%, wherein the percentages refer to the mass percentages of the component in the raw materials for the first auxiliary alloy; or

the raw material for the first auxiliary alloy is composed of the following components: Pr with a content of 7.5%; Nd with a content of 22.5%; Ho with a content of 20%; Cu with a content of 5%; Co with a content of 5%; Fe with a content of 39.5%; and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the components in the raw material for the first auxiliary alloy.

4. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 1, wherein:

in S1, the RH_2 accounts for 50-80% by mass, for example, 58% by mass or 63% by mass of the second auxiliary alloy sheet; and/or

in S1, the X_2 accounts for 6-10% by mass of the second auxiliary alloy sheet; and/or

in S1, when the X_2 comprises Zr, the Zr accounts for 6-10% by mass of the second auxiliary alloy sheet; and/or

in S1, the raw material for the second auxiliary alloy sheet further comprises LH_2 , the LH_2 is one or more of La, Ce, Pr, and Nd, and the LH_2 accounts for 80% by mass or less, excluding 0% by mass of the second auxiliary alloy sheet; and/or

in S1, the raw material for the second auxiliary alloy sheet further comprises B, and in the second auxiliary alloy sheet, the B accounts for 0-0.6% by mass, excluding 0% by mass, for example, 0.5% by mass of the second auxiliary alloy sheet.

5. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 4, wherein:

the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 58%; Zr with a content of 6%; and Fe with a content of

36%, wherein the percentages refer to the mass percentages of the components in the raw material for the second auxiliary alloy; or

the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 63%; Zr with a content of 6%; Fe with a content of 30.5%;
5 and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the components in the raw material for the second auxiliary alloy; or

the raw material for the second auxiliary alloy is composed of the following components: Dy with a content of 58%; Zr with a content of 6%; Fe with a content of 35.5%;
and B with a content of 0.5%, wherein the percentages refer to the mass percentages of the
10 components in the raw material for the second auxiliary alloy.

6. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 1, wherein:

in S1, the first auxiliary alloy sheet has a melting point which is lower than that of the
15 second auxiliary alloy sheet.

7. The preparation method of the double-shell neodymium-iron-boron magnet according to claim 1, wherein:

the LH_3 accounts for 27-30% by mass, for example, 27.6% by mass, 28% by mass or
20 29% by mass of the main alloy sheet; and/or

the X_3 accounts for 0.1-0.6% by mass, preferably 0.2-0.3% by mass, for example 0.22%
by mass of the main alloy sheet; and/or

the Y_3 accounts for 0.75-2.5% by mass, for example 1.15% by mass or 2.06% by mass
of the main alloy sheet; and/or

25 in the main alloy sheet, the Cu accounts for 0.1-0.6% by mass, for example 0.21% by mass or 0.25% by mass of the main alloy sheet; and/or

in the main alloy sheet, when the Y_3 comprises Al, the Al accounts for 0.02-1.2% by mass, for example 0.4% by mass of the main alloy sheet; and/or

in the main alloy sheet, when the Y_3 comprises Ga, the Ga accounts for 0.25-0.4% by
30 mass, for example 0.26% by mass of the main alloy sheet; and/or

in the main alloy sheet, when the Y_3 comprises Co, the Co accounts for 0.5-1.2% by mass, for example 1.19% by mass of the main alloy sheet; and/or

in the main alloy sheet, the B accounts for 0.97-1% by mass of the main alloy sheet; and/or

5 the raw material for the main alloy sheet further comprises RH_3 , the RH_3 is Dy and/or Tb, and the RH_3 accounts for 0-3% by mass, excluding 0% by mass of the main alloy sheet;

preferably, the raw material for the main alloy is composed of the following components: Nd with a content of 27.6%; Dy with a content of 3%; Ga with a content of 0.26%; Al with a content of 0.4%; Cu with a content of 0.21%; Co with a content of 1.19%;
10 Ti with a content of 0.2%; Nb with a content of 0.02%; B with a content of 1%; Fe with a content of 66.12%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy; or

the raw material for the main alloy is composed of the following components: Pr with a content of 7%; Nd with a content of 21%; Dy with a content of 3%; Ga with a content of
15 0.4%; Cu with a content of 0.25%; Co with a content of 0.5%; Zr with a content of 0.2%; B with a content of 0.97%; and Fe with a content of 66.68%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy; or

the raw material for the main alloy is composed of the following components: Pr with a content of 7.25%; Nd with a content of 21.75%; Dy with a content of 1%; Ga with a content
20 of 0.4%; Cu with a content of 0.25%; Co with a content of 0.5%; Zr with a content of 0.2%; B with a content of 0.97%; Fe with a content of 67.68%, wherein the percentages refer to the mass percentages of the components in the raw material for the main alloy.

8. The preparation method of the double-shell neodymium-iron-boron magnet
25 according to claim 1, wherein:

in S1, the first auxiliary alloy sheet is obtained by smelting and casting the raw material for the first auxiliary alloy sheet; preferably, the temperature for smelting the raw material for the first auxiliary alloy sheet is 1000°C or more; the casting for the first auxiliary alloy sheet is preferably a strip continuous casting process, an ingot casting process, a
30 centrifugal casting process, or a rapid quenching process; and/or

in S1, the second auxiliary alloy sheet is obtained by smelting and casting the raw material for the second auxiliary alloy sheet; preferably, the temperature for smelting the raw material for the second auxiliary alloy sheet is 1300°C or more; the casting for the second auxiliary alloy sheet is preferably a strip continuous casting process, an ingot casting process,
5 a centrifugal casting process, or a rapid quenching process; and/or

in S1, the main alloy sheet is obtained by smelting and casting the raw material for the main alloy sheet; preferably, the temperature for smelting the raw material for the main alloy sheet is 1400°C or more; the casting for the main alloy sheet is preferably a strip continuous casting process, an ingot casting process, a centrifugal casting process, or a rapid quenching
10 process; and/or

in S2, the amount of the first auxiliary alloy sheet and the second auxiliary alloy sheet accounts for more than 1% by mass and less than 15% by mass, preferably 5% by mass of the amount of the raw material for the double-shell neodymium-iron-boron magnet; the raw material for the double-shell neodymium-iron-boron magnet comprises the main alloy sheet,
15 the first auxiliary alloy sheet and the second auxiliary alloy sheet; and/or

in S2, a mixture of the main alloy sheet and the auxiliary alloy sheets is subjected to hydrogen decrepitation, pulverization, shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet; or, the main alloy sheet and the auxiliary alloy sheets are subjected to hydrogen decrepitation respectively, then the coarse powders obtained after
20 hydrogen decrepitating the main alloy sheet and the auxiliary alloy sheets are mixed, and then the mixed coarse powders are subjected to pulverization, shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet; or, the main alloy sheet and the auxiliary alloy sheets are subjected to hydrogen decrepitation and pulverization respectively, then the coarse powders obtained after pulverizing the main alloy sheet and the auxiliary alloy
25 sheets are mixed, and then the mixed coarse powders are subjected to shaping and sintering treatments to obtain the double-shell neodymium-iron-boron magnet; and/or

in S2, the hydrogen decrepitation has a dehydrogenation temperature of 400°C-650°C, for example, 500-620°C; and/or

in S2, the pulverization is jet mill pulverization; and/or

30 in S2, the pulverization is carried out under an atmosphere with an oxygen content of

50 ppm or less; and/or

in S2, the particles after the pulverization have a diameter of 2-7 μm , for example, 3.0-5.3 μm ; and/or

in S2, the shaping comprises pressing into a green compact in a press with a magnetic
5 field strength of 0.5T-3.0T, for example, 1.0-2.0T; and/or

in S2, the sintering temperature is 1000-1150°C, for example, 1050-1085°C.

9. A double-shell neodymium-iron-boron magnet obtainable by a method of any of the
previous claims, comprising a main phase crystal grain, a double shell of the main phase
10 crystal grain and an Nd-rich phase adjacent to the main phase crystal grain,

the main phase crystal grain comprises $\text{R}_2\text{Fe}_{14}\text{B}$, wherein the R comprises Nd and
comprises one or more of La, Ce, and Pr;

the double shell has an inner layer comprising $(\text{Nd}/\text{Ho})_2\text{Fe}_{14}\text{B}$ and/or $(\text{Nd}/\text{Gd})_2\text{Fe}_{14}\text{B}$;

the double shell has an outer layer comprising $(\text{Nd}/\text{Dy})_2\text{Fe}_{14}\text{B}$ and/or $(\text{Nd}/\text{Tb})_2\text{Fe}_{14}\text{B}$;

15 the double shell has a thickness of 0.1-6 μm ;

the Nd-rich phase comprises a $(\text{R}-\text{RH})_6\text{T}_{13}\text{X}$ phase, wherein RH is one or more of Ho,
Gd, Dy and Tb; T is Fe and/or Co; and X is one or more of Ga, Cu and Al.

10. The double-shell neodymium-iron-boron magnet according to claim 9, wherein
20 the Nd-rich phase further comprises ZrB_2 and TiB_2 .