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(54) RARE EARTH SOFT MAGNETIC POWDER AND PREPARATION METHOD THEREFOR, AND SOFT MAGNETIC COMPOSITE MATERIAL AND PREPARATION METHOD THEREFOR

(57) Disclosed are rare-earth soft magnetic powder and a preparation method therefor, and a soft magnetic composite material and a preparation method therefor The rare-earth soft magnetic powder comprises $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, wherein T comprises any one or a combination of at least two of Si, C, or B; x, y, and z are atomic contents of Ni, T, and N, respectively, wherein x ranges from 0.1 to 0.5, y ranges from 0.1 to 0.5, and z ranges from 2 to 4; the soft magnetic composite material is prepared by mixing the rare-earth soft magnetic powder with a binder, which can meet the use of electronic devices under a high-frequency working condition.

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Description

TECHNICAL FIELD

[0001] Embodiments of the present application relate to the technical field of magnetic materials, for example, rare-earth soft magnetic powder and a preparation method therefor, and a soft magnetic composite material and a preparation method therefor.

BACKGROUND

[0002] In recent years, with the development of miniaturization, multi-functionality, rapid processing speed, and high drive frequency of mobile communication devices (computers, mobile phones, digital cameras, and car navigation systems, etc.), the industrial sector has an increasing demand for soft magnetic materials able to have a low loss characteristic in variation of high-frequency electromagnetic field and ultra-high-frequency electromagnetic field.

[0003] At present, the common soft magnetic materials on the market comprise ferrite soft magnetic materials (manganese-zinc, and nickel-zinc), metal soft magnetic materials (Fe, Fe-Si, Fe-Si-Al, Fe-Si-Cr, and Fe-Ni, etc.), and amorphous nanocrystalline soft magnetic materials. The ferrite soft magnetic material can work at a frequency of 1 MHz due to low eddy current loss at a high-frequency working condition lead by high resistivity. However, the saturation magnetization of the ferrite soft magnetic material is low (< 0.5 T), which is not suitable for the preparation of very small magnetic components. Compared with the ferrite soft magnetic material, the metal soft magnetic material has a higher saturation magnetization which makes it suitable for the miniaturization of magnetic devices. However, the metal soft magnetic material has small resistivity, which leads to a very large eddy current loss under a high-frequency working condition, resulting in great reduction of the energy utilization efficiency, as well as shortened working lifetime caused by the generate heat violently of the magnetic devices, so the working frequency of the metal soft magnetic material generally does not exceed 100 kHz. The amorphous nanocrystalline soft magnetic material has higher saturation magnetization than the ferrite soft magnetic material, and has high resistivity at the same time, and has a fine grain size, thus the amorphous nanocrystalline soft magnetic material can have a relatively small eddy current loss under a high-frequency working condition. Thereby, the amorphous nanocrystalline soft magnetic material can work at a higher electromagnetic field frequency, for example, nano-soft magnetic powder particles treated with a good insulation coating can even work at a frequency of 100 MHz.

[0004] In recent years, it has been found that easy-plane rare-earth-iron-nitrogen compounds (such as Ce-Fe-N, and Nd-Fe-N) have a very high cut-off frequency, reaching 6 GHz, which exceeds the cut-off frequency of microwave ferrites. However, the resistance of the materials is much lower than that of ferrite materials, which leads to a very large eddy current loss under a high-frequency working condition, thus the high-frequency characteristics of the easy-plane rare-earth-iron-nitrogen compounds are difficult to be fully utilized.

[0005] CN114974786A discloses a soft magnetic composite material, which comprises a first soft magnetic metal powder, a first insulation coating material, and jet milling-granulating powder. The jet milling-granulating powder comprises second soft magnetic metal powder and a second insulation coating material, and the second insulation coating material is completely cured. The first insulation coating material and/or the second insulation coating material in the soft magnetic composite material are evenly dispersed. The soft magnetic composite material is used to prepare a metal powder core and a molded inductor, and the prepared molded inductor has high insulation resistance and high initial magnetic permeability.

[0006] CN114023522A discloses a micron-scale magnetic composite material with good stability. The magnetic material comprises metal compound magnetic particles and micro-nano magnetic composite particles, wherein the micro-nano magnetic composite particles are iron particles, and the iron particles contain silica particles with an average powder particle size of 10 nm, and the metal compound magnetic particles comprise a composition of Fe-Si-based soft magnetic alloy particles, iron-aluminum-based soft magnetic alloy particles, iron-silicon-aluminum-based soft magnetic alloy particles, iron-chromium-based soft magnetic alloy particles, nickel-based soft magnetic alloy particles, and two-dimensional magnetic-moment micro-powder. The micron-scale composite magnetic material prepared by the method has a high doping concentration of the magnetic particles, and has the advantages of simple preparation processes and high stability. Meanwhile, due to the coating effect of the macromolecule organic matter, the prepared magnetic polymer micro-nano composite particles also have good biocompatibility.

[0007] CN113724958A discloses a preparation method for producing an iron-based soft magnetic iron core based on the alloying of reduced iron powder, and belongs to the field of soft magnetic materials and powder metallurgy technology. The method comprises: (1) reducing reduced iron powder in a high purity with hydrogen; (2) mixing high-purity reduced iron powder with alloy powder and a binder evenly, then subjecting to alloying to obtain iron-based soft magnetic powder; (3) performing a surface inorganic insulation coating treatment; (4) adding the iron-based soft magnetic powder after the coating treatment into an acetone resin solution, heating and stirring, and then mixing the same with a release agent evenly for pressing to obtain an iron-based soft magnetic composite block; and (5) subjecting the iron-based soft magnetic composite block to a heat treatment to obtain an iron-based soft magnetic iron core. The

method has a simple process and a low cost, which can effectively reduce a burning loss of the alloy component during the melting process, and basically eliminates defects such as satellite powder and surface pits, and avoids a case where composition segregation is easy to occur during the solidification process.

[0008] However, the above soft magnetic materials have a high loss in a high-frequency electromagnetic field or an ultra-high-frequency electromagnetic field, thus their popularization and application are limited to a certain extent. Therefore, it is of great significance to develop rare-earth soft magnetic powder and a preparation method therefor as well as a soft magnetic composite material and a preparation method therefor to which can meet the use of electronic devices under a GHz working condition.

SUMMARY

[0009] The following is a summary of the subject described herein. This summary is not intended to limit the protection scope of the claims.

[0010] Embodiments of the present application provide rare-earth soft magnetic powder and a preparation method therefor, and a soft magnetic composite material and a preparation method therefor. Various raw material powder is mixed, and then subjected to a heat treatment, a vacuum heat treatment, a melting treatment, and a rapid cooling treatment, and the temperature of the melting treatment is strictly controlled to obtain a mixture in a glassy state; the mixture in the glassy state is heated in a nitrogen atmosphere, and undergo a crystallization reaction and a nitriding reaction to obtain the rare-earth soft magnetic powder finally; the soft magnetic composite material is prepared by mixing the rare-earth soft magnetic powder with a binder, which has low eddy current loss at high-frequency and can meet the use of electronic devices under a high-frequency working condition.

[0011] In a first aspect, an embodiment of the present application provides rare-earth soft magnetic powder, and the rare-earth soft magnetic powder comprises $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, wherein T comprises any one or a combination of at least two of Si, C, or B; x, y, and z are atomic contents of Ni, T, and N, respectively, wherein x ranges from 0.1 to 0.5, y ranges from 0.1 to 0.5, and z ranges from 2 to 4.

[0012] The rare-earth soft magnetic powder in the present application comprises $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ having an easy-plane structure, wherein Ce is a key substance of constituting the $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ compound, and an appropriate addition of ferromagnetic element Ni can improve the soft magnetic characteristics of the material such as magnetic permeability; and T comprises any one or a combination of at least two of Si, C, or B, and by adding an appropriate amount of the above substances, an eddy current loss of the material at a high-frequency can be reduced, but the excessive additive amount will reduce the soft magnetic characteristics of the material. Each element is combined according to a specific atomic ratio and acts synergistically, and the soft magnetic composite material prepared from the formed rare-earth soft magnetic powder can be applied to a high-frequency environment of more than or equal to 1 GHz, and has low eddy current loss.

[0013] In the present application, x ranges from 0.1 to 0.5, which may be, for example, 0.1, 0.2, 0.3, 0.4, or 0.5, etc., but the x is not limited to the listed values, and other unlisted values within the numerical range are also applicable; y ranges from 0.1 to 0.5, which may be, for example, 0.1, 0.2, 0.3, 0.4, or 0.5, etc., but the y is not limited to the listed values, and other unlisted values within the numerical range are also applicable; and z ranges from 2 to 4, which may be, for example, 2, 2.5, 3, 3.5, 3.8, or 4, but the z is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0014] Preferably, a shape of the rare-earth soft magnetic powder comprises a lamellar shape.

[0015] Preferably, the rare-earth soft magnetic powder has an average particle size of 50-100 nm, which may be, for example, 50 nm, 60 nm, 70 nm, 80 nm, 90 nm, or 100 nm, etc.; however, the average particle size is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0016] In a second aspect, an embodiment of the present application further provides a preparation method for the rare-earth soft magnetic powder according to the first aspect, and the preparation method comprises the following steps:

(1) mixing Fe_2O_3 powder, Ce_2O_3 powder, Ni_2O_3 powder, SiO_2 powder, C powder, and B_2O_3 powder as raw materials according to $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ to obtain mixed powder;

(2) subjecting the mixed powder to a heat treatment in a hydrogen atmosphere, then mixing the same with a reducing agent and an additive, and performing a vacuum heat treatment to obtain an intermediate product with a loose texture;

(3) subjecting the intermediate product to a melting treatment and a rapid cooling treatment in sequence, and then heating in a nitrogen atmosphere to undergo a crystallization reaction and a nitriding reaction to obtain a post-reaction substance; and the heating is performed at a temperature of 400-550 °C; and

(4) mixing the post-reaction substance and an acetic acid solution, performing stirring and solid-liquid separation in sequence to obtain the rare-earth soft magnetic powder.

[0017] In the preparation method for the rare-earth soft magnetic powder in the present application, the mixed powder is subjected to a heat treatment in the hydrogen atmosphere which aims to obtain ultrafine metal Fe pow-

der via reducing Fe_2O_3 in the mixed powder; then the high activity of the reducing agent is utilized to reduce Ce_2O_3 to obtain rare-earth metal Ce; the additive is utilized to reduce the melting temperature of the raw material mixture participating in the vacuum heat treatment to obtain the intermediate product with a loose texture; then the intermediate product is sequentially subjected to a melting treatment and a rapid cooling treatment to form the mixture in a glassy state; the mixture in the glassy state is heated in a nitrogen atmosphere to undergo a crystallization reaction and a nitriding reaction to obtain the post-reaction substance; finally, a non-magnetic substance in the post-reaction substance is dissolved into a solution by using the acetic acid solution, and the rare-earth soft magnetic powder is obtained in the end.

[0018] The mixture in the glassy state in the present application is heated at a temperature of 400-550 °C in a nitrogen atmosphere. In a case where the heating temperature is high, the post-reaction substance will not form an easy-plane structure with a phase composition of $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, and a large number of α-Fe phases will appear. In a case where the heating temperature is low, the final obtained rare-earth soft magnetic powder will have a low nitrogen content and significantly increased magnetic loss.

[0019] The heating in the present application is performed at a temperature of 400-550 °C, which may be, for example, 400 °C, 420 °C, 450 °C, 500 °C, 530 °C, or 550 °C, etc.; however, the temperature is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0020] Preferably, the heat treatment in step (2) is performed at a temperature of 700 °C.

[0021] Preferably, the heat treatment is performed for a period of 10 h.

[0022] Preferably, the hydrogen atmosphere has a hydrogen purity of more than 99.9%, which may be, for example, 99.9%, 99.91%, 99.93%, 99.95%, 99.97%, or 99.99%, etc.; however, the hydrogen purity is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0023] Preferably, the reducing agent comprises metal calcium.

[0024] Preferably, the additive comprises B_2O_3 powder.

[0025] In the present application, the additive preferably comprises B_2O_3 powder, and its effect is mainly to reduce the melting temperature of the raw material mixture participating in the vacuum heat treatment, and the addition of the B_2O_3 powder does not affect the composition of the final $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, i.e., the T in the $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ is any one or a combination of at least two of Si, C, or B, which is determined by the original raw materials (SiO_2 powder, C powder, or B_2O_3 powder).

[0026] Preferably, an additive amount of the reducing agent accounts for 15% of a mass of the mixed powder.

[0027] Preferably, an additive amount of the additive accounts for 3% of a mass of the mixed powder.

[0028] Preferably, the vacuum heat treatment in step (2) has a vacuum degree of 10^{-2} Pa.

[0029] Preferably, a heating step of the vacuum heat treatment comprises: first heating to 800 °C, and then introducing argon, and heating to 1000 °C.

[0030] Preferably, the vacuum heat treatment is performed with a temperature-holding period of 4 h.

[0031] Preferably, the vacuum heating treatment is followed by cooling to 20-30 °C, which may be, for example, 20 °C, 23 °C, 25 °C, 27 °C, 29 °C, or 30 °C, etc.; however, the temperature is not limited to the listed values, and other unlisted values within the numerical range are also applicable

[0032] Preferably, the melting treatment in step (3) is performed at a temperature of 1300-1500 °C, which may be, for example, 1300 °C, 1350 °C, 1400 °C, 1450 °C, 1470 °C, or 1500 °C, etc.; however, the temperature is not limited to the listed values, and other unlisted values within the numerical range are also applicable

[0033] The temperature of the melting treatment is preferably 1300-1500 °C, and the intermediate product with a loose texture is subjected to the melting treatment and rapid cooling treatment to form a mixture in the glassy state. In a case where the temperature of the melting treatment is low, the diffusion rate of each element in the material will be low, the distribution of each component in the alloy will be uneven, and the proportion of the soft magnetic phase in the alloy will be reduced, and the soft magnetic composite prepared from the rare-earth soft magnetic powder will have a low magnetic loss, but the magnetic permeability will be greatly reduced. In a case where the temperature of the melting treatment is high, more energy consumption will be generated, and a preparation cost of the rare-earth soft magnetic powder will be increased.

[0034] Preferably, the rapid cooling treatment is achieved by the following method: a molten liquid after the melting treatment is flowed to a molybdenum roller rotating at a high speed through a quartz nozzle so as to cool the same rapidly to form a mixture in the glassy state.

[0035] Preferably, the molybdenum roller has a rotation speed of 20-25 m/s, which may be, for example, 30 m/s, 21 m/s, 22 m/s, 23 m/s, 24 m/s, or 25 m/s, etc.; however, the rotation speed is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0036] Preferably, the heating is performed for a period of 6-10 h, which may be, for example, 6 h, 7 h, 8 h, 9 h, or 10 h, etc.; however, the period is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0037] In step (4) of the present application, the operation step of mixing the post-reaction substance and the acetic acid solution can be that the post-reaction substance is firstly immersed in deionized water, then added with the acetic acid solution, and continuously stirred, so that the non-magnetic substance in the post-reaction substance can be dissolved in water. In the present ap-

plication, there is no need to clearly limits a concentration of the acetic acid solution, as long as ensures that non-magnetic impurities such as CaO in the reactive substance can be completely dissolved, and then the solid-liquid separation treatment is performed. The operation in step (4) is repeated until a content of Ca ions in the reactive substance is less than 0.1%.

[0038] Preferably, the solid-liquid separation comprises filtration.

[0039] As a preferred technical solution of the present application, the preparation method comprises the following steps:

(1) mixing Fe_2O_3 powder, Ce_2O_3 powder, Ni_2O_3 powder, SiO_2 powder, C powder, and B_2O_3 powder as raw materials according to $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ to obtain mixed powder;

(2) subjecting the mixed powder to a heat treatment at a temperature of 700 °C for 10 h in a hydrogen atmosphere with a purity of more than 99.9%, then mixing the same with a reducing agent and an additive, and performing a vacuum heat treatment with a vacuum degree of 10^{-2} Pa for 4 h to obtain an intermediate product with a loose texture;
the reducing agent comprises metal calcium; the additive comprises B_2O_3 powder; an additive amount of the reducing agent accounts for 15% of a mass of the mixed powder; an additive amount of the additive accounts for 3% of a mass of the mixed powder; a heating step of the vacuum heat treatment comprises: first heating to 800 °C, and then introducing argon, and heating to 1000 °C; and the vacuum heating treatment is followed by cooling to 20-30 °C;;

(3) subjecting the intermediate product to a melting treatment at a temperature of 1300-1500 °C and a rapid cooling treatment in sequence, and then heating at a temperature of 400-550 °C for 6-10 h in a nitrogen atmosphere to undergo a crystallization reaction and a nitriding reaction to obtain a post-reaction substance;
the rapid cooling treatment is achieved by a method below: flowing a molten liquid after the melting treatment to a high-rotated molybdenum roller at a rotation speed of 20-25 m/s through a quartz nozzle so as to cool the same rapidly to form a mixture in the glassy state; and

(4) mixing the post-reaction substance and an acetic acid solution, performing stirring and solid-liquid separation in sequence to obtain the rare-earth soft magnetic powder.

[0040] In a third aspect, an embodiment of the present application further provides a soft magnetic composite material, and the soft magnetic composite material is prepared by mixing the rare-earth soft magnetic powder according to the first aspect with a binder; the rare-earth soft magnetic powder accounts for 20-70% of a mass of the soft magnetic composite material, which may be, for example, 20%, 30%, 40%, 50%, 60%, or 70%, etc.; however, the proportion is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0041] The soft magnetic composite material in the present application is prepared by mixing the rare-earth soft magnetic powder according to the first aspect with a binder; the binder comprises a thermosetting binder such as an epoxy resin, and also comprises a thermoplastic binder such as Nylon 6, Nylon 12, or polyphenylene sulfide, etc. The rare-earth soft magnetic powder accounts for 20-70% of a mass of the soft magnetic composite material in the present application; in a case where the proportion of the rare-earth soft magnetic powder is less than 20%, the magnetic permeability of the soft magnetic composite material will be low, which is not conducive to the miniaturization of magnetic devices; in a case where the proportion of the rare-earth soft magnetic powder is more than 70%, the loss of the soft magnetic composite material at a high-frequency will be significantly increased.

[0042] In a fourth aspect, an embodiment of the present application also provides a preparation method for the soft magnetic composite material according to the third aspect, and the preparation method comprises the following steps:

(a) mixing an epoxy resin and acetone to obtain a mixed solution;

(b) mixing the mixed solution with rare-earth soft magnetic powder, and stirring to obtain rare-earth soft magnetic powder coated with an epoxy resin; and

(c) drying the rare-earth soft magnetic powder coated with an epoxy resin, then mixing with a thermoplastic binder, blending and granulating to obtain the soft magnetic composite material.

[0043] In the preparation method for the soft magnetic composite material in the present application, the rare-earth soft magnetic powder is firstly mixed with a mixed solution containing an epoxy resin to coat a layer of a macromolecule resin on the surface of the rare-earth soft magnetic powder to form a protective layer, so that in the subsequent process of blending and granulating, the rare-earth soft magnetic powder is not easy to be oxidized.

[0044] Preferably, an additive amount of the epoxy resin in step (a) accounts for 3% of a mass of the rare-earth soft magnetic powder.

[0045] Preferably, a volume ratio of the mixed solution and the rare-earth soft magnetic powder is 1.5:1-2:1, which may be, for example, 1.5:1, 1.6:1, 1.7:1, 1.8:1, or

2:1, etc.; however, the volume ratio is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0046] Preferably, the drying in step (c) is performed at temperature of 70 °C.

[0047] Preferably, the drying is performed for a period of 1 h.

[0048] Preferably, the thermoplastic binder comprises any one or a combination of at least two of Nylon 6, Nylon 12, or polyphenylene sulfide, wherein a typical but non-limiting combination comprises a combination of Nylon 6 and Nylon 12, a combination of polyphenylene sulfide and Nylon 6, or a combination of Nylon 12, polyphenylene sulfide, and Nylon 6.

[0049] Preferably, an additive amount of the thermoplastic binder accounts for 17-67% of a mass of the rare-earth soft magnetic powder coated with an epoxy resin, which may be, for example, 17%, 20%, 30%, 50%, 60%, or 67%, etc.; however, the proportion is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0050] Preferably, the blending is performed at a temperature of 5-10 °C higher than a softening temperature of the thermoplastic binder, which may be, for example, 5 °C, 6 °C, 7 °C, 8 °C, 9 °C, or 10 °C, etc.; however, the temperature difference is not limited to the listed values, and other unlisted values within the numerical range are also applicable.

[0051] Preferably, argon is introduced for protection during the blending.

[0052] As a preferred technical solution of the present application, the preparation method comprises the following steps:

(a) mixing an epoxy resin and acetone to obtain a mixed solution; mixing the mixed solution with rare-earth soft magnetic powder, and stirring to obtain rare-earth soft magnetic powder coated with an epoxy resin;
an additive amount of the epoxy resin accounts for 3% of a mass of the rare-earth soft magnetic powder;
a volume ratio of the mixed solution and the rare-earth soft magnetic powder is 1.5:1-2:1; and

(b) drying the rare-earth soft magnetic powder coated with an epoxy resin at a temperature of 70 °C for 1 h, and then mixing the same with a thermoplastic binder, and performing blending and granulating to obtain the soft magnetic composite material;

the thermoplastic binder comprises any one or a combination of at least two of Nylon 6, Nylon 12, or polyphenylene sulfide; an additive amount of the thermoplastic binder accounts for 17-67% of a mass of the rare-earth soft magnetic powder coated with an epoxy resin; the blending is performed at a temperature of 5-10 °C higher than a softening temperature of the thermoplastic binder; argon is introduced for protection during the blending.

[0053] Compared with the related art, the embodiments in the present application have the following beneficial effects at least:

(1) in the preparation method for the rare-earth soft magnetic powder provided in the embodiments of the present application, the rare-earth soft magnetic powder with a fine particle size is prepared, and the soft magnetic composite material prepared from the rare-earth soft magnetic powder can be applied to an environment with a high-frequency of more than or equal to 1 GHz, and has a low eddy current loss; and

(2) the preparation method for the rare-earth soft magnetic powder and the preparation method for the soft magnetic composite material provided in the examples of the present application have a simple process and a low preparation cost, which have a prospect of large-scale popularization and application.

[0054] Other aspects will be appreciated upon reading and understanding the detailed description.

DETAILED DESCRIPTION

[0055] To facilitate understanding the present application, examples are listed below in the present application. Those skilled in the art should understand that the examples merely assist in understanding the present application but should not be regarded as a specific limitation of the present application.

[0056] The present application is explained in further detail below. However, the following examples are only brief examples of the present application, and do not represent or limit the protection scope of the claims in the present application. The protection scope in the present application is subject to the claims.

Example 1

[0057] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method comprises the following steps:

(1) Fe_2O_3 powder, Ce_2O_3 powder, Ni_2O_3 powder, SiO_2 powder, C powder, and B_2O_3 powder as raw materials were mixed according to $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ to obtain a mixed powder, wherein x = 0.2, y = 0.2, and T was element Si;

(2) the mixed powder was put into a tube furnace, subjected to a heat treatment at a temperature of 700 °C for 10 h in a hydrogen atmosphere with a purity of more than 99.9%, then mixed with metal calcium as a reducing agent and B_2O_3 powder as an additive, and subjected to a vacuum heat treat-

ment with a vacuum degree of 10^{-2} Pa for 4 h to obtain an intermediate product with a loose texture; an additive amount of the reducing agent accounted for 15% of a mass of the mixed powder; an additive amount of the additive accounted for 3% of the mass of the mixed powder; a heating step of the vacuum heat treatment comprised: first, the temperature was raised to 800 °C, and then argon was introduced, and the temperature was raised to 1000 °C; and the vacuum heating treatment was followed by cooling to 20-30 °C;

(3) the intermediate product was put into a melting furnace, subjected to a melting treatment at a temperature of 1300 °C to melt the same completely, and then the molten liquid flowed to a high-rotated molybdenum roller with a rotation speed of 20 m/s through a quartz nozzle for a rapid cooling treatment to form a mixture in a glassy state; the mixture in the glassy state was placed in a treatment furnace with a nitrogen atmosphere, and heated at a temperature of 500 °C for 6 h to undergo a crystallization reaction and a nitriding reaction to obtain a post-reaction substance; and

(4) the post-reaction substance was immersed in deionized water, and then added with an acetic acid solution, and stirred and filtered sequentially to obtain the rare-earth soft magnetic powder.

[0058] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.6}Ni_{0.2}Si_{0.2}N_{2.8}$ with an average particle size of 80 nm.

[0059] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method comprises the following steps:

(a) an epoxy resin and acetone was mixed to obtain a mixed solution; the mixed solution and the rare-earth soft magnetic powder were mixed and stirred to obtain rare-earth soft magnetic powder coated with an epoxy resin;
an additive amount of the epoxy resin accounted for 3% of a mass of the rare-earth soft magnetic powder; a volume ratio of the mixed solution and the rare-earth soft magnetic powder was 1.5:1; and

(b) the rare-earth soft magnetic powder coated with an epoxy resin was dried at a temperature of 70 °C for 1 h, and then mixed with a thermoplastic binder Nylon 6, and then subjected to blending and granulating in a twin-screw granulator to obtain the soft magnetic composite material;

an additive amount of the thermoplastic binder accounted for 40% of the mass of the rare-earth soft magnetic powder coated with the epoxy resin; the blending was performed at a temperature of 8 °C higher than a softening temperature of the thermoplastic binder; argon was introduced for protection during the blending.

[0060] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 4.22 and magnetic loss $\tan\delta\mu$ was 0.11. **Example 2**

[0061] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that in step (1), x = 0.1, and y = 0.1.

[0062] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.8}Ni_{0.1}Si_{0.1}N_{2.8}$ with an average particle size of 82 nm.

[0063] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0064] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 4.33 and magnetic loss $\tan\delta\mu$ was 0.13. **Example 3**

[0065] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that in step (1), x = 0.5, and y = 0.5.

[0066] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.0}Ni_{0.5}Si_{0.5}N_{2.8}$ with an average particle size of 75 nm.

[0067] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0068] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 3.75 and magnetic loss $\tan\delta\mu$ was 0.10. **Example 4**

[0069] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that the melting treatment in step (3) was performed at a temperature of 1500 °C

[0070] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.6}Ni_{0.2}Si_{0.2}N_{2.7}$ with an average particle size of 85 nm.

[0071] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft

magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0072] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 4.25 and magnetic loss tan$\delta\mu$ was0.13.

Example 5

[0073] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that the heating in step (3) was performed at a temperature of 400 °C for a period of 10 h.

[0074] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.6}Ni_{0.2}Si_{0.2}N_{2.5}$ with an average particle size of 55 nm.

[0075] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0076] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 3.83 and magnetic loss tan$\delta\mu$ was 0.07. **Example 6**

[0077] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that the heating in step (3) was performed at a temperature of 550 °C for a period of 6 h.

[0078] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.6}Ni_{0.2}Si_{0.2}N_{3.2}$ with an average particle size of 100 nm.

[0079] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0080] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 4.83 and magnetic loss tan$\delta\mu$ was 0.13.

Example 7

[0081] This example provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the rare-earth soft magnetic powder in Example 1 with a binder. The preparation method is the same as that of Example 1 except that an additive amount of the thermoplastic binder Nylon 6 accounted for 17% of a mass of the rare-earth soft magnetic powder coated with an epoxy resin.

[0082] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 6.30 and magnetic loss tan$\delta\mu$ was 0.21.

Example 8

[0083] This example provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the rare-earth soft magnetic powder in Example 1 with a binder. The preparation method is the same as that of Example 1 except that an additive amount of the thermoplastic binder Nylon 6 accounted for 67% of a mass of the rare-earth soft magnetic powder coated with an epoxy resin.

[0084] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 2.18 and magnetic loss tan$\delta\mu$ was 0.03.

Example 9

[0085] This example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that SiO_2 powder was replaced by C powder, and T was element C in the raw material of step (1).

[0086] A composition of the rare-earth soft magnetic powder obtained in this example was $Ce_2Fe_{16.6}Ni_{0.2}C_{0.2}N_{2.8}$ with an average particle size of 82 nm.

[0087] This example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0088] The soft magnetic composite material obtained in this example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz. The measured magnetic permeability μ' was 4.38 and magnetic loss tan$\delta\mu$ was 0.11.

Comparative Example 1

[0089] This comparative example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that the heating in step (3) was performed at a temperature of 700 °C for a period of 10 h.

[0090] The rare-earth soft magnetic powder obtained in this comparative example did not form an easy-plane structure with a phase composition of $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, and a large number of α-Fe phases appeared in the rare-earth soft magnetic powder.

Comparative Example 2

[0091] This comparative example provides a preparation method for rare-earth soft magnetic powder, and the preparation method is the same as in Example 1 except that the heating in step (3) was performed at a temperature of 350 °C for a period of 10 h.

[0092] A composition of the rare-earth soft magnetic powder obtained in this comparative example was $Ce_2Fe_{16.6}Ni_{0.2}C_{0.2}N_{1.1}$ with an average particle size of 41 nm.

[0093] This comparative example also provides a soft magnetic composite material, and the soft magnetic composite material was prepared by mixing the above rare-earth soft magnetic powder with a binder. The preparation method is the same as that of Example 1.

[0094] The soft magnetic composite material obtained in this comparative example was prepared into a circular ring sample, and tested with a vector network analyzer at a frequency of 1 GHz, and the magnetic permeability μ' was 4.08 and magnetic loss tan$\delta\mu$ was 0.28. The soft magnetic composite obtained in this comparative example had a low nitrogen content and a significantly increased magnetic loss.

[0095] In summary, the preparation method for the rare-earth soft magnetic powder and the preparation method for the soft magnetic composite material provided in the present application have a simple process and a low preparation cost. The final obtained soft magnetic composite material is suitable for use in a high-frequency working condition, and especially, able to meet the use of electronic devices under a GHz working condition.

[0096] The applicant declares that the above is only the embodiments of the present application, but the protection scope of the present application is not limited thereto. Those skilled in the art should understand that any change or replacement, which can be easily thought of by a person skilled in the art within the scope of the technology disclosed in the present application, shall fall within the protection scope and disclosure scope of the present application.

Claims

1. Rare-earth soft magnetic powder, which comprises $Ce_2Fe_{17-x-y}Ni_xT_yN_z$, wherein T comprises any one or a combination of at least two of Si, C, or B; x, y, and z are atomic contents of Ni, T, and N, respectively, wherein x ranges from 0.1 to 0.5, y ranges from 0.1 to 0.5, and z ranges from 2 to 4.

2. The rare-earth soft magnetic powder according to claim 1, wherein a shape of the rare-earth soft magnetic powder comprises a lamellar shape.

3. The rare-earth soft magnetic powder according to claim 1 or 2, wherein the rare-earth soft magnetic powder has an average particle size of 50-100 nm.

4. A preparation method for the rare-earth soft magnetic powder according to any one of claims 1-3, comprising the following steps:

   (1) mixing Fe_2O_3 powder, Ce_2O_3 powder, Ni_2O_3 powder, SiO_2 powder, C powder, and B_2O_3 powder as raw materials according to $Ce_2Fe_{17-x-y}Ni_xT_yN_z$ to obtain mixed powder;
   (2) subjecting the mixed powder to a heat treatment in a hydrogen atmosphere, then mixing the same with a reducing agent and an additive, and performing a vacuum heat treatment to obtain an intermediate product with a loose texture;
   (3) subjecting the intermediate product to a melting treatment and a rapid cooling treatment in sequence, and then heating in a nitrogen atmosphere to undergo a crystallization reaction and a nitriding reaction to obtain a post-reaction substance; and the heating is performed at a temperature of 400-550 °C; and
   (4) mixing the post-reaction substance and an acetic acid solution, performing stirring and solid-liquid separation in sequence to obtain the rare-earth soft magnetic powder.

5. The preparation method according to claim 4, wherein the hydrogen atmosphere in step (2) has a hydrogen purity of more than 99.9%.

6. The preparation method according to claim 4 or 5, wherein the reducing agent comprises metal calcium.

7. The preparation method according to any one of claims 4-6, wherein the additive comprises B_2O_3 powder.

8. The preparation method according to any one of claims 4-7, wherein a heating step of the vacuum heat treatment in step (2) comprises: heating to 800 °C, then introducing argon, and heating to 1000 °C; preferably, the vacuum heating treatment is followed by cooling to 20-30 °C.

9. The preparation method according to any one of claims 4-8, wherein the melting treatment in step (3) is performed at a temperature of 1300-1500 °C;

   preferably, the rapid cooling treatment is achieved by the following method: flowing a molten liquid after the melting treatment to a molybdenum roller rotating at a high speed through a quartz nozzle so as to cool the same rapidly to form a mixture in the glassy state;
   preferably, the molybdenum roller has a rotation speed of 20-25 m/s;

preferably, the heating is performed for a period of 6-10 h.

10. A soft magnetic composite material, which is prepared by mixing the rare-earth soft magnetic powder according to any one of claims 1-3 with a binder; and the rare-earth soft magnetic powder accounts for 20-70% of a mass of the soft magnetic composite material.

11. A preparation method for the soft magnetic composite material according to claim 10, comprising the following steps:

(a) mixing an epoxy resin and acetone to obtain a mixed solution; mixing the mixed solution with rare-earth soft magnetic powder, and stirring to obtain rare-earth soft magnetic powder coated with an epoxy resin; and
(b) drying the rare-earth soft magnetic powder coated with an epoxy resin, then mixing the same with a thermoplastic binder, and blending and granulating to obtain the soft magnetic composite material.

12. The preparation method according to claim 11, wherein a volume ratio of the mixed solution and the rare-earth soft magnetic powder is 1.5: 1-2:1.

13. The preparation method according to claim 11 or 12, wherein the thermoplastic binder comprises any one or a combination of at least two of Nylon 6, Nylon 12, or polyphenylene sulfide.

14. The preparation method according to any one of claims 11-13, wherein an additive amount of the thermoplastic binder accounts for 17-67% of a mass of the rare-earth soft magnetic powder coated with an epoxy resin.

15. The preparation method according to any one of claims 11-14, wherein the blending is performed at a temperature of 5-10 °C higher than a softening temperature of the thermoplastic binder;
preferably, argon is introduced for protection during the blending.

INTERNATIONAL SEARCH REPORT

International application No.
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A. CLASSIFICATION OF SUBJECT MATTER

H01F1/153(2006.01)i; H01F41/02(2006.01)i; H01F1/147(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: H01F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

CNTXT, ENTXT, ENTXTC, DWPI, CNKI: 稀土, 铈, 铁, 氮, 磁, 镍, 硅, 碳, 硼, 掺杂, 熔炼, 快冷, 速冷, 快速冷却, 快淬, rare, earth, Re, Ce, Fe, N, magnetic, Ni, Si, C, B, doping, doped, smelt+, rapid, cool+, quench+

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
PX	CN 115497704 A (HENGDIAN GROUP DMEGC MAGNETICS CO., LTD.) 20 December 2022 (2022-12-20) description, paragraphs 0009-0087	1-15
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☑ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
07 September 2023	**13 September 2023**
Name and mailing address of the ISA/CN **China National Intellectual Property Administration (ISA/CN)** **China No. 6, Xitucheng Road, Jimenqiao, Haidian District, Beijing 100088**	Authorized officer Telephone No.

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INTERNATIONAL SEARCH REPORT

International application No.
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INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2023/097141

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REFERENCES CITED IN THE DESCRIPTION

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