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METHOD FOR PRODUCING INTERMETALLIC COMPOUND PERMANENT MAGNET MATERIAL

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ABSTRACT OF THE DISCLOSURE

An intermetallic compound for fine particle permanent magnets is produced by mixing a rare earth metal hydride powder with a purified 3d-metal powder in a stoichiometric ratio under a protective gas atmosphere, the mixture is then pressed to form a tablet, and the tablet is then sintered at a temperature of from 750 to 1250 deg. C. for a period of from four to ten hours under exclusion of nitrogen, oxygen and carbon. The preferred 3d-metal is cobalt sponge and suitable rare earth metals from which the metal hydride is formed include samarium and yttrium.

This invention relates to an improved method for producing an intermetallic compound for fine particle permanent magnets, from inter-metallic compounds consisting of at least one 3d-transition element and one or more rare earth metals.

It is known to produce fine particle permanent magnets, by means of inter-metallic compounds of binary systems of a 3d-transition element and a rare earth metal. Preferably, binary compounds of the group R_2Co_{17} and RCo_5 in combination with the rare earth metals Y, La, Ce, Pr, Nd or Sm have been used. Inter-metallic compounds for fine particle magnets of the order of Microns (μ) were produced by grinding blanks of an inter-metallic compound obtained by fusion metallurgy.

This known method has the disadvantage that the magnetic properties of the binary compounds of the 3d-transition element and the rare earth are changed in an unfavorable manner during the grinding step. Up to a particle size of from 10 to 15 μ the coercive force increases and then drops as the grinding is continued. This deterioration of the magnetic properties is known as the "overgrinding" effect.

The principal object of the present invention is to provide an improved method for producing an inter-metallic compound for fine particle permanent magnets in which this unfavorable, overgrinding effect is avoided.

The problem is solved, according to the present invention, in that a rare earth metal hydride powder is mixed with a purified 3d metal powder in a stoichiometric ratio according to the desired inter-metallic compound in a protective gas atmosphere, then pressed to a blank, and then sintered at a temperature of from 750 to 1250 deg. C. for a period of from four to ten hours under exclusion of nitrogen, oxygen and carbon.

The improved method, according to the invention, yields an easily disintegratable sintered body which requires very little grinding. A further advantage is that the sintering temperature is far below the respective melting temperatures of the components. Consequently, there is no reaction with the material from which the sintering crucible is made, which thereby results in a substantially purer and better magnet material. The rare earth metal hydride can be formed by heating under a vacuum the rare earth metal at a temperature of 600 deg. C. for several hours to remove oxygen and nitrogen that may still be absorbed on

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the surface after a preliminary mechanical cleaning process, followed by passage of hydrogen at a temperature of 300 deg. C. until complete saturation is achieved.

The mechanical cleaning process for liberation of the oxide and nitride coats before the heating process can be effected in an inert nitrogen flow.

Preferably, the passage of hydrogen is effected during at least one hour.

After the rare earth metal has been converted to hydride form, it can be ground under a protective atmosphere of argon.

The 3d-metal, preferably cobalt, is mixed preferably as a sponge with the rare earth metal hydride powder and then pressed under a pressure of 200 kp./cm.² to form a blank.

The sintering operation of the blank can take place in a vacuum or under a protective gas atmosphere.

The improved method of producing the fine particle magnets has been carried out in accordance with the following examples.

EXAMPLE 1**(a) Production of Sm-hydride**

A bright piece of samarium metal (1.0 g.), mechanically cleaned under streaming nitrogen, was heated under a vacuum at a temperature of 600 deg. C. for three hours in order to remove oxygen and nitrogen that may still be adsorbed on the surface after the cleaning. In the same apparatus, well purified hydrogen was then conducted over the metallic piece at 300 deg. C. until complete saturation was achieved. This required a reaction time of about two hours. The SmH_3 thus formed was stored under hydrogen gas.

(b) Production of Co_5Sm

The samarium hydride which had been stored was now ground, in a cupola box filled with argon, to about micron size and then mixed mechanically with cobalt sponge, the weighed-in ratio being 1000 g. of SmH_3 to 1960 g. Co. This mixture was then pressed under a pressure of 200 kp./cm.² to form a tablet having a diameter of 13 mm. The tablet was then sintered for ten hours at a temperature of 950 deg. C. under a vacuum of 2.10^{-8} torr.

The product was then identified by X-ray crystallography as Co_5Sm .

A portion of the sintered tablet was powdered, without grinding, by mechanical crushing and examined in a magnetometer. The coercive force measured 5000 oe.

EXAMPLE 2**(a) Production of Y-hydride**

A bright piece of yttrium metal (4.0 g.), mechanically cleaned under streaming nitrogen, was heated under a vacuum at a temperature of 600 deg. C. for four hours in order to remove oxygen and nitrogen that might still be adsorbed on the surface after the cleaning. In the same apparatus, well-purified hydrogen was then conducted over the metallic piece at 400 deg. C. until complete saturation was achieved. This required a reaction time of eight hours. The reaction product was identified by X-ray crystallography as YH_3 , and was then stored in a polished vessel under hydrogen gas.

(b) Production of Co_5Y

The yttrium hydride which had been stored was now ground in a cupola box filled with argon, to about micron size and then mixed mechanically with cobalt sponge, the weighed-in ratio being 4.00 g. of YH_3 to 13.10 g. of Co. This mixture was then pressed under a pressure of 200 kp./cm.² to form several tablets having a diameter of 13

mm. and a height of from 8 to 10 mm. The tablets were then sintered for eight hours at a temperature of 1180 deg. C. under a vacuum of 10^{-4} torr.

The inter-metallic compound produced was identified by X-ray crystallography as Co_5Y . A powder produced by mechanical crushing yielded a coercive force of 1650 oe.

EXAMPLE 3

(a) Production of Sm-hydride

A bright piece of samarium metal (12.0 g.), mechanically cleaned under streaming nitrogen, was heated under a vacuum at a temperature of 600 deg. C. for five hours in order to remove oxygen and nitrogen that might still be adsorbed on the surface after the cleaning. In the same apparatus, well-purified hydrogen was then conducted over the metallic piece at 300 deg. C. until complete saturation was achieved. The reaction time with the hydrogen was five hours. The reaction product was identified by X-ray crystallography as SmH_3 , and was then stored in a polished vessel under hydrogen gas.

(b) Production of Co_5Sm

The samarium hydride which had been stored was now ground in a cupola box filled with argon, to about micron size and then mixed with cobalt sponge, the weighed-in ratio being 12.00 g. of SmH_3 to 23.51 of Co. This mixture was then pressed under a pressure of 200 kp./cm.² to form several tablets having a diameter of 13 mm. and a height of from 6 to 7 mm. The tablets were then sintered for five hours at a temperature of 980 deg. C. under a vacuum of 2.10^{-6} torr.

The inter-metallic compound produced was identified by X-ray crystallography as Co_5Sm . A powder produced from the tablets by mechanical crushing yielded a coercive field strength of from 6500 to 8000 oe.

We claim:

1. The method for producing an intermetallic compound of a 3d-transition metal and at least one rare earth metal for fine particle permanent magnets which comprises the steps of mixing at least one purified rare earth metal hydride powder with a purified 3d-metal powder in a stoichiometric ratio according to the desired intermetallic compound under a protective gas atmosphere, pressing said powder mixture into a tablet, and then sintering said tablet at a temperature of from 750° to 1250° C. for a period of from four to ten hours under exclusion of nitrogen, oxygen and carbon.

2. The method as defined in claim 1, wherein said rare earth metal hydride is formed by the steps which include heating the rare earth metal in a vacuum at a temperature of 600° C. for a time sufficient to liberate any oxide or nitride that may still be adsorbed on the surface, and then passing hydrogen in contact with the purified rare earth metal at a temperature of 300° C. until complete saturation is achieved.

3. The method as defined in claim 2 which includes the further step of mechanically cleaning the rare earth metal in a nitrogen flow before it is heated to liberate said oxide or nitride that may still be adsorbed.

4. The method as defined in claim 2 wherein the hydrogen is passed in contact with the purified rare earth metal for at least one hour.

5. The method as defined in claim 1, wherein the rare earth metal hydride is ground to substantially micron size under a protective argon atmosphere prior to mixing with the 3d-metal powder.

6. The method as defined in claim 1, wherein the rare earth metal is Y, La, Ce, Pr, Nd or Sm.

7. The method as defined in claim 1, wherein the 3d-metal is cobalt.

8. The method as defined in claim 7, wherein the 3d-metal is cobalt sponge.

9. The method as defined in claim 1, wherein the sintering step is carried out in a vacuum.

10. The method as defined in claim 1, wherein the sintering step is carried out under a protective gas atmosphere.

11. The method as defined in claim 1, wherein the powder mixture is pressed to tablet form under a pressure of 200 kp./cm.².

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