

Hard Magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ Compound by Mechanical Alloying

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Abstract

In the present work, mechanical alloying technique was applied to prepare hard magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders. Starting from pure Fe and Sm powders, the formation process of hard magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ phase by mechanical alloying and subsequent solid-gas reaction was studied. At as milled condition powders were found to consist of Sm-Fe amorphous and α -Fe phases in all compositions of $\text{Sm}_x\text{Fe}_{100-x}$ ($x = 11, 13, 15, 17$). The effects of starting composition on the formation of $\text{Sm}_2\text{Fe}_{17}$ intermetallic compound was investigated by heat treatment of mechanically alloyed powders. When Sm content was 15 at. %, heat-treated powders consisted of nearly $\text{Sm}_2\text{Fe}_{17}$ single phase. For preparation of hard magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders, additional nitriding treatment was performed under N_2 gas flow at 450 °C. The increase in the coercivity was proportional to the nitrogen content, which increased drastically at first and then increased gradually as the nitriding time was extended to 3 h.

INTRODUCTION

Coey *et al.* have shown that the magnetic properties of a $\text{Sm}_2\text{Fe}_{17}$ intermetallic compound can be changed dramatically to a magnetically hard one by an incorporation of nitrogen atoms into its rhombohedral $\text{Th}_2\text{Zn}_{17}$ lattice [1]. The intrinsic magnetic properties of thus obtained $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ interstitial compound are very excellent: the material has a high Curie temperature T_C , a high saturation magnetization M_s , and an extremely high value of the uniaxial magnetic anisotropy H_a [2]. All these properties are superior even to the well-known Nd-Fe-B type permanent magnet. Therefore magnets of Sm-Fe-N should have the potential to compete favourably with the Nd-Fe-B magnets.

The $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ nitride is usually prepared by conventional melting and subsequent solid-gas reaction between $\text{Sm}_2\text{Fe}_{17}$ alloy and nitrogen at an elevated temperature [1, 3]. In general, we realize that one of the difficulties in pre-

paring nitride material is the reactivity of mother alloy with nitrogen gas. For synthesizing $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ compound, therefore, the $\text{Sm}_2\text{Fe}_{17}$ alloy is processed into powder material in order to enhance the reactivity. Mechanical alloying (MA) has been recognized as a new technique for synthesizing permanent magnetic powder materials through solid-state diffusion under intense mechanical deformations [4–6]. In particular, it has been shown that the mechanical alloying process may prove to be a useful and cost effective method for the production of isotropic polycrystalline permanent magnetic powder materials [4, 5].

In the present work, mechanical alloying technique was applied to prepare permanent magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders. Starting from pure Fe and Sm powders, the formation process of hard magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ phase by mechanical alloying was studied. In addition, the effects of starting composition on the formation of $\text{Sm}_2\text{Fe}_{17}$ intermetallic compound was inves-

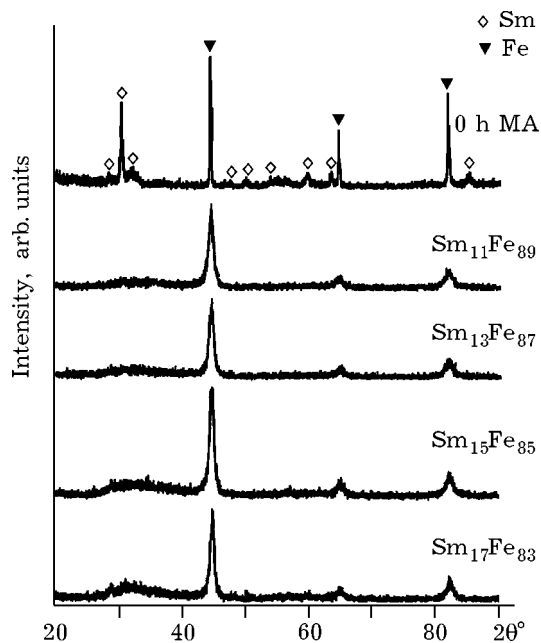


Fig. 1. X-ray diffraction patterns of $\text{Sm}_x\text{Fe}_{100-x}$ ($x = 11, 13, 15, 17$) powders mechanically alloyed for 40 h.

tigated by heat treatment of mechanically alloyed powders. The change in magnetic properties of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ phase was also studied as a function of nitrogen content.

EXPERIMENTAL

The mechanical alloying was carried out at room temperature for a mixture of pure iron (99.9 %, <150 μm in size) and pure samarium (99.9 %, <750 μm in size) powders with a composition $\text{Sm}_x\text{Fe}_{100-x}$ ($x = 11, 13, 15, 17$) in an Ar (99.999 %) gas atmosphere. A planetary ball mill (Fritsch Pulverisette 5) was used with its vial rotation of 200 rpm. The vial and balls are made of the hardened steel (SKD11). The total mass of powders was about 20 g and the ratio of balls to powders was 7 : 1. Milling was interrupted for 6 min in every 30 min to suppress the excessive temperature rise of the vial and balls. The mechanically alloyed powder was heat treated at 700 °C for 40 min in vacuum, and subsequent nitriding treatment at 450 °C was performed for various time intervals.

The structural change of ball-milled powders was studied by X-ray diffraction in continuous and step scanning modes with $\text{CuK}\alpha$ radiation. The morphology of the particles was observed using the scanning electron microscope.

The magnetization at room temperature was measured with the vibrating sample magnetometer calibrated using a pure Ni disk (99.99 %). The nitrogen content absorbed into the sample was determined with a nitrogen-oxygen analyzer.

RESULTS AND DISCUSSION

The X-ray diffraction patterns are shown in Fig. 1 as a function of Sm content in $\text{Sm}_x\text{Fe}_{100-x}$ ($x = 11, 13, 15, 17$) powders obtained by MA treatment for 40 h. It can be seen that a two-phase mixture of amorphous Sm-Fe and a-Fe was formed in all compositions of MA powders. The broadening of the a-Fe diffraction lines is caused by the refinement in grains and also by the accumulation of defects and strains.

Figure 2 shows the X-ray diffraction patterns when the as-milled powder was heat treated at 700 °C for 40 min in vacuum. When Sm content is 15 at. %, it is found that the as-milled powder is transformed to nearly single-phase $\text{Sm}_2\text{Fe}_{17}$ compound with rhombohedral $\text{Th}_2\text{Zn}_{17}$ crystal structure. Only a small amount of Sm_2O_3 oxide is also observed. The Sm content

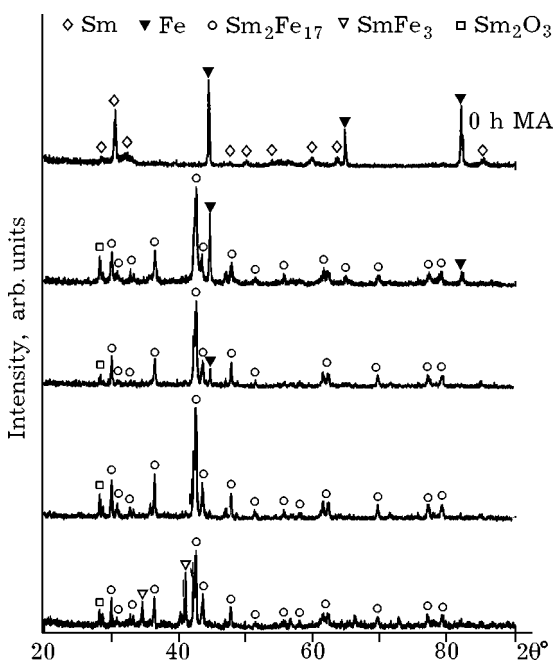


Fig. 2. X-ray diffraction patterns of $\text{Sm}_x\text{Fe}_{100-x}$ ($x = 11, 13, 15, 17$) MA powders after the formation of $\text{Sm}_2\text{Fe}_{17}$ phase by annealing at 700 °C for 40 min in vacuum.

in $\text{Sm}_{15}\text{Fe}_{85}$ powder is higher than that required for the formation of nominal 2 : 17 phase. We believe that this is deeply related to the possible Sm losses during heat treatment due to the high Sm vapour pressure or additional $\alpha\text{-Fe}$ by wear debris during intense milling. In this state, the powder material is still magnetically soft because of the magnetic in-plane anisotropy of $\text{Sm}_2\text{Fe}_{17}$ phase.

The change in the morphology of MA powders was studied as a function of milling time. Figure 3 shows the scanning electron micrographs for the $\text{Sm}_{15}\text{Fe}_{85}$ powders treated by MA 40 h

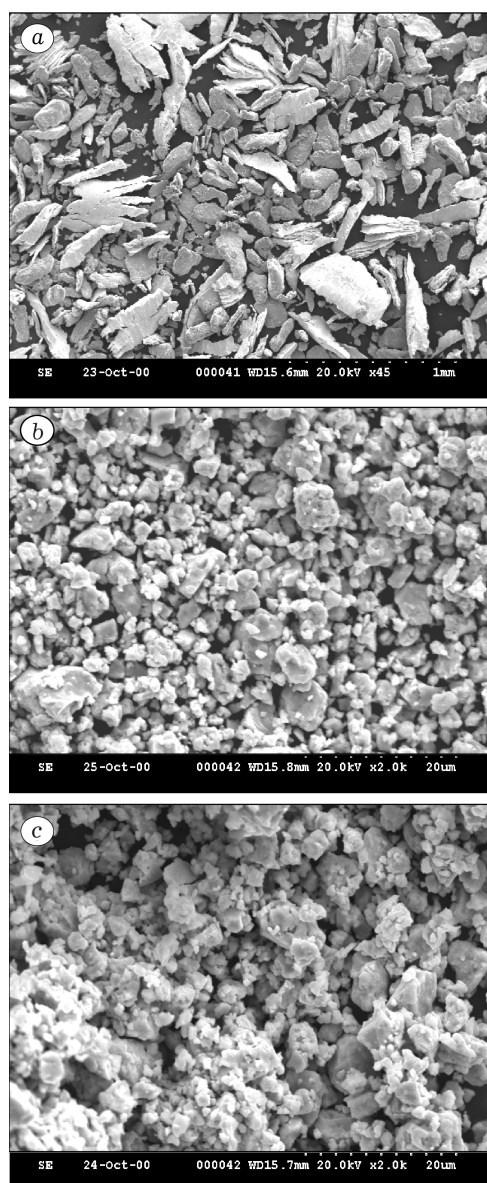


Fig. 3. SEM micrographs of $\text{Sm}_{15}\text{Fe}_{85}$ powders for initial sample (a), 40 h MA (b) and nitrided at 450 °C for 3 h (c).

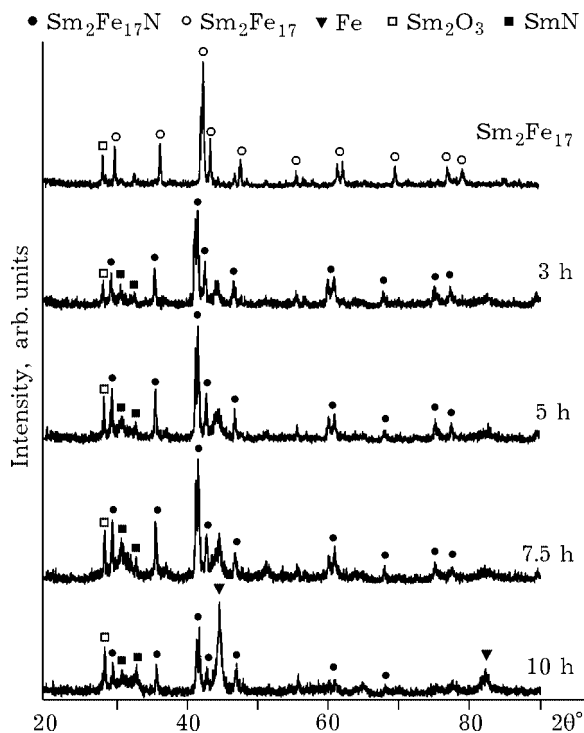


Fig. 4. X-ray diffraction patterns of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders nitrided at 450 °C for various time intervals.

and that of subsequently nitrided at 450 °C for 3 h. The powder of MA 40 h is fairly round and its size distribution is centered at about 2 mm diameter. In contrast, the nitrided powder is more irregular and agglomerate and its size distribution is much broader.

For magnetic hardening the subsequent nitriding treatment is required, where the nitrogen atoms are incorporated into the rhombohedral $\text{Th}_2\text{Zn}_{17}$ crystal structure, which can be monitored by an expanding of the lattice parameter. Figure 4 shows the X-ray diffraction patterns of $\text{Sm}_2\text{Fe}_{17}$ compound nitrided at 450 °C for various time intervals. The X-ray diffraction patterns indicate a slight shift of the $\text{Sm}_2\text{Fe}_{17}$ lines to low diffraction angle, suggesting that there is no structural change of the $\text{Th}_2\text{Zn}_{17}$ crystal structure, but lattice parameter has been expanded. The nitrogen content increased drastically for 3 h and then decreased gradually as nitriding time was extended. As is clear from in Fig. 4, the decrease in nitrogen content after 3 h of nitrogenation is related to the decomposition of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ nitride into $\alpha\text{-Fe}$ and Sm_2O_3 . It is also noted that lattice parameters of nitrides are constant irrespective of nitrogen content, suggesting

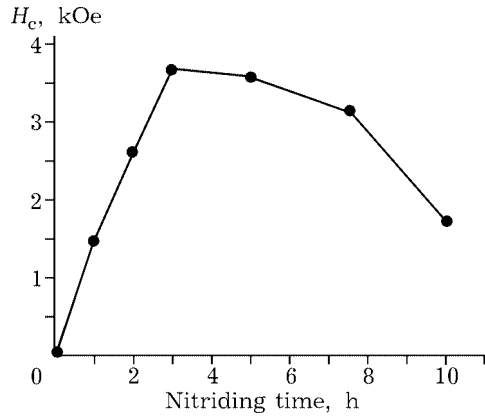


Fig. 5. Coercivity of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders nitrided at $450\text{ }^\circ\text{C}$ for various time intervals.

that the nitriding time affects mainly the amount of nitride.

The coercivity of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ powders nitrided at $450\text{ }^\circ\text{C}$ for various time intervals is shown in Fig. 5. The increase in the coercivity is proportional to the nitrogen content, which increased drastically at first and then increased gradually as the nitriding time was extended to 3 h. In contrast, the low coercivity after 3 h of nitrogenation is apparently due to the precipitation of soft magnetic α -Fe phase, as recognized from Fig. 4.

CONCLUSIONS

The hard magnetic $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ nitride powder may be obtained, by mechanical alloying of a mixture of pure Sm and pure Fe powders with a composition $\text{Sm}_{15}\text{Fe}_{85}$ and subsequent solid-gas reaction in N_2 atmosphere. The increase in the coercivity is proportional to the nitrogen content as the nitriding time is extended to 3 h.

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