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LETTER TO THE EDITOR

## A hard magnetic property study of a novel $\text{Sm}_3(\text{Fe}, \text{Ti})_{29}\text{N}_y$ nitride

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**Abstract.** The hard magnetic properties of a new interstitial nitride  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  ( $y \sim 5$ ) have been investigated by ball milling. The saturation magnetization linearly decreases with ball-milling time. By ball milling, maxima of remanence  $B_r = 1.04$  T, intrinsic coercivity  $\mu_{0i}H_c = 0.83$  T and energy product  $(BH)_{\text{max}} = 105$  kJ m<sup>-3</sup> are achieved at 293 K.

Recently new ternary phases of  $\text{Nd}_2(\text{Fe}, \text{Ti})_{19}$  [1] and  $\text{R}_2(\text{Fe}_{0.91}\text{V}_{0.09})_{17}$  ( $\text{R}=\text{Y}, \text{Nd}, \text{Sm}, \text{Gd}$ ) have been discovered [2, 3]. The crystal structure of this new phase has been identified from x-ray diffraction to be an  $\text{Nd}_3(\text{Fe}, \text{Ti})_{29}$ -type structure by Li *et al* [4]. The symmetry is monoclinic (space group  $P2_1/C$ ). The substitution fractions of dumbbell sites in  $\text{CaCu}_5$ -type structure are  $\frac{1}{3}$ ,  $\frac{2}{3}$  and  $\frac{1}{2}$  for  $\text{Th}_2\text{Zn}_{17}$  type,  $\text{Nd}_3(\text{Fe}, \text{Ti})_{29}$  type and  $\text{ThMn}_{12}$  type respectively. The magnetic properties of these 3:29 compounds have also been reported [1-3, 5-7]. The nitrogenation effect on  $\text{R}_2(\text{Fe}_{0.91}\text{V}_{0.09})_{17}$  compounds has been investigated by Shcherbakova *et al* [3]; it is similar to those on 2:17 and 1:12 systems. The interstitial phase  $\text{R}_3(\text{Fe}, \text{Ti})_{29}\text{N}_{4-\delta}$  was determined by Li *et al* based on the consideration of the structure relationship [8]:  $\text{R}_3(\text{Fe}, \text{Ti})_{29} = \text{R}_2\text{Fe}_{17} + \text{R}(\text{Fe}, \text{Ti})_{12}$ . The interstitial N atoms are distributed over two crystallographic sites: namely,  $4e_1(\frac{1}{3}, \frac{1}{2}, \frac{2}{3})$  and  $4e_2(\frac{1}{2}, \frac{1}{4}, \frac{1}{4})$ . In our previous work, we reported the formation and intrinsic magnetic properties of  $\text{Sm}_3(\text{Fe}, \text{Ti})_{29}$  [9] and its nitride  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  ( $y \sim 5$ ) [10]. The  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  nitride has almost the same intrinsic magnetic properties as  $\text{Sm}_2\text{Fe}_{17}\text{N}_y$  [11], which indicates that the former is also a potential candidate for permanent magnet application. In this work, we investigated the hard magnetic properties of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  by the conventional ball-milling technique. The results are encouraging for the development of permanent magnet material.

The  $\text{Sm}_{10}\text{Fe}_{84}\text{Ti}_6$  alloy was prepared by Ar arc melting starting elements with purity of at least 99.9%, followed by annealing in vacuum at a temperature of 1273-1473 K for 10-60 h, wrapping in Mo foil and sealing in quartz tubes, then quenching in water. An excess amount of Sm was added to compensate the loss of the Sm during the melting. The x-ray diffraction pattern shows that the  $\text{Sm}_{10}\text{Fe}_{84}\text{Ti}_6$  is a single phase of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}$  with the same crystalline structure as  $\text{Nd}_3(\text{Fe}, \text{Ti})_{29}$  [10]. The alloy was pulverized into fine powder with an average particle size of 15  $\mu\text{m}$ . The nitrogenation was performed by heating the fine powder in  $\text{N}_2$  at 1 atm at a temperature of 770-870 K for about 2 h.

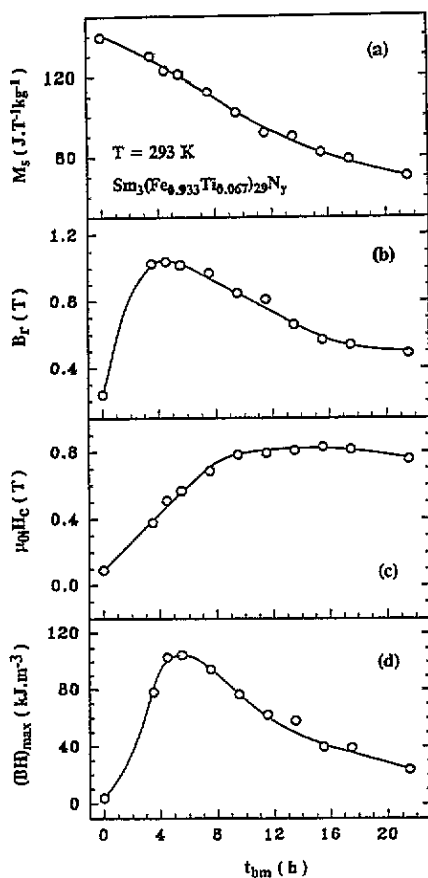


Figure 1. The variation of hard magnetic properties of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder with ball-milling time.

The N concentration is about five N atoms per 3:29 formula, obtained from the mass difference between before and after nitrogenation, which corresponds to three N atoms per 2:17 formula. The  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  nitride is also a single phase with the same structure as that of the parent compound, except for a little  $\alpha$ -Fe inside. After nitrogenation the relative volume expansion is about 7.1%, which is similar to those found in 2:17 nitrides [11]. The nitride powder then was ball milled using 4–10 mm diameter metal balls with a weight ratio of 1:60 between the sample and the metal balls. The sample for the magnetic measurements was made by mixing the ball-milled nitride powder with epoxy resin, solidifying in an applied field of 2 T.

X-ray diffraction with Cu  $K\alpha$  radiation and thermomagnetic analysis performed in a low field of 0.04–0.08 T were used to identify the phases. The hysteresis loops were measured by means of a pulsed magnetic field and an SQUID up to 5 T.

As the ball-milling time  $t_{\text{bm}}$  increases, the saturation magnetization  $M_s$  of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder decreases linearly (see figure 1(a)). The value of  $M_s$  decreases to half of the value for the non-milled powder when  $t_{\text{bm}} = 21.5$  h. The reason is that, by ball milling, the grains are broken and the crystalline structure in the surface of the particles is destroyed, so the main phase of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  decreases while the non-magnetic impurity appears. This was observed in the x-ray diffraction patterns with

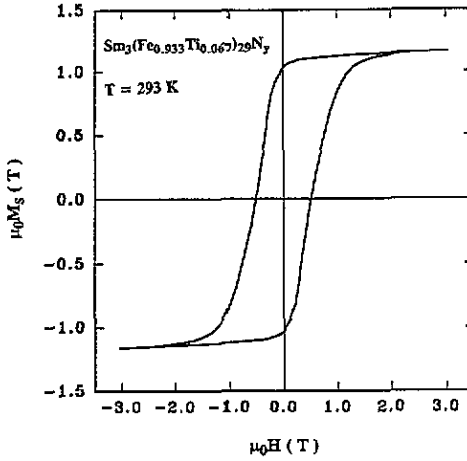


Figure 2. The hysteresis loop of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder with a ball-milling time of 4.5 h at 293 K.

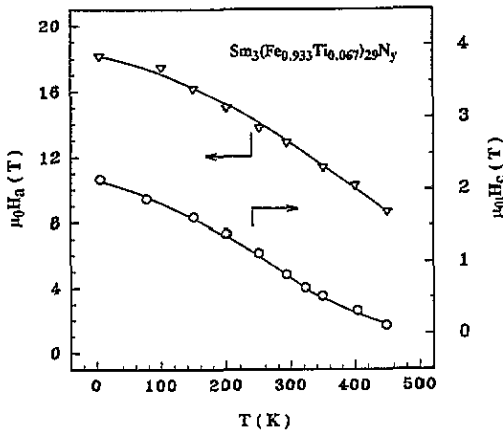


Figure 3. The temperature dependence of the coercivity of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder with a ball-milling time of 11.5 h and the anisotropy field of the  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  compound.

different ball-milling time  $t_{\text{bm}}$ ; the longer the  $t_{\text{bm}}$  the worse the pattern.

The remanence  $B_r$  of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder increases rapidly at the beginning of the ball milling, reaches a maximum of 1.04 T at  $t_{\text{bm}} = 4.5$  h and then decreases slowly (see figure 1(b)). The intrinsic coercivity  $\mu_{01}H_c$  has a different feature from the remanence (see figure 1(c)). With increasing ball-milling time  $t_{\text{bm}}$ ,  $\mu_{01}H_c$  increases almost linearly until  $t_{\text{bm}} = 10$  h, reaches a plateau of about 0.82 T between  $t_{\text{bm}} = 10$  h and 16 h, and then decreases very slowly. The variation of energy product  $(BH)_{\text{max}}$  is similar to that of  $B_r$  (see figure 1(d)). The maximum value of  $(BH)_{\text{max}}$  is  $105 \text{ kJ m}^{-3}$  when  $t_{\text{bm}} = 4.5$  h, at which  $B_r$  has the maximum value.

Figure 2 shows the hysteresis loop at 293 K of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder ball milled for 4.5 h. For the sample with  $t_{\text{bm}} < 3$  h,  $B_r$  is too small to give a high enough value of  $(BH)_{\text{max}}$ , while for the sample with long ball-milling time,  $(BH)_{\text{max}}$  has small values

for two reasons: the small  $B_r$  values and poor squareness of the hysteresis loops.

The temperature dependence of the coercivity  $\mu_{0i}H_c$  of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  powder for  $t_{\text{bm}} = 11.5$  h compared with the anisotropy field  $\mu_0H_a$  is shown in figure 3. The coercivity only reaches  $\frac{1}{9}$  and  $\frac{1}{15}$  of the anisotropy field at 4.2 K and 293 K respectively. On plotting  $\mu_{0i}H_c/\mu_0M_s$  versus  $\mu_{0i}H_a/\mu_0M_s$  at different temperatures, the relationship  $\mu_{0i}H_c = c\mu_{0i}H_a - N_eM_s$  holds quite well, where  $N_e$  is the effective demagnetization factor and  $c$  is related to the alignment of the grains and the surface layer properties of the grains. Therefore, for a very fine powder of  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$ , it is more possible to have nucleation located at the inhomogeneous surface layer of the main phase of the grains rather than pinning, which is similar to the observation on explosion sintering  $\text{Sm}_2\text{Fe}_{17}\text{N}_y$  magnets [12, 13] and in  $\text{NdFe}_{12-x}\text{Mo}_x\text{N}_y$  nitride [14]. A detailed discussion of the coercivity mechanism will be published separately [15].

In conclusion, the hard magnetic properties of the new  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  nitride have been studied by ball milling. Values of coercivity  $\mu_{0i}H_c = 0.83$  T and energy product  $(BH)_{\text{max}} = 105$  kJ m<sup>-3</sup> at 293 K are obtained. These data, as initial results of the hard magnetic property study, show that  $\text{Sm}_3(\text{Fe}_{0.933}\text{Ti}_{0.067})_{29}\text{N}_y$  is suitable for development for permanent magnet application.

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