# ARPN Journal of Engineering and Applied Sciences

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### RESEARCH OF THE EFFECT OF Sm<sub>2</sub>Fe<sub>17</sub> ALLOYING WITH TITANIUM AND MOLYBDENUM ON MAGNETIC PROPERTIES

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#### ABSTRACT

Development of high Nd<sub>2</sub>Fe<sub>14</sub>B based energy-intensive magnets has shown that in principle R-Fe intermetallic compounds with high content of iron are good and relatively cheap magnetic materials. However, a disadvantage of such compounds is the low Curie temperature, which decreases in the R-Fe system with increasing of iron concentration, and becomes the smallest ~ 300-400 K in the case of R<sub>2</sub>Fe<sub>17</sub>. Magnetic properties of the compound can be significantly modified by doping of various interstitial or substitutional impurities. The work presents the results of the investigation on the effect of Sm<sub>2</sub>Fe<sub>17</sub> alloying with titanium and molybdenum, in particular, on the Curie temperature (T<sub>C</sub>) and magnetic properties. It is shown that Sm<sub>2</sub>Fe<sub>17</sub> alloying with titanium and molybdenum increases the lattice parameter and the volume of the lattice cell without changing the lattice symmetry. When measuring the hysteretic properties of Sm<sub>2</sub>Fe<sub>17</sub> based alloys, it was revealed that the introduction of titanium, as well as molybdenum, leads to a broadening of the hysteresis loop.

Keywords: Sm<sub>2</sub>Fe<sub>17</sub>, energy-intensive magnets, magnetic materials, magnetic properties, hysteretic properties.

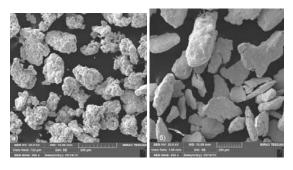
### 1. INTRODUCTION

In recent years, the interest in the research of magnetism of R<sub>2</sub>Fe<sub>17</sub> and R(Fe<sub>1</sub>T)<sub>12</sub> type compounds (where T = Ti, V, Mo, W, Cr, Si) with high iron content has increased dramatically due to obtaining on their basis of new compounds with small atoms of light interstitial elements, such as nitrogen and carbon with magnetic ordering temperatures and the effective magnetic anisotropy fields, exceeding the proper values for Nd<sub>2</sub>Fe<sub>14</sub>B compounds [1-11]. Creation of high Nd<sub>2</sub>Fe<sub>14</sub>B based energy-intensive magnets has shown that, in principle, R-Fe intermetallic compounds with high content of iron are good and relatively cheap magnetic materials [12, 13]. However, a disadvantage of such compounds is low Curie temperature, which in the R-Fe system decreases with increasing of iron concentration and becomes the smallest  $\sim 300\text{-}400 \text{ K}$  in the case of  $R_2Fe_{17}$ containing the largest amount of iron [5, 6, 14]. Magnetic properties of the compound can be significantly modified by the introduction of various interstitial or substitutional impurities [14-19]. These substitutions may lead to an improvement of the magnetic properties of Sm<sub>2</sub>Fe<sub>17</sub> compounds in the context of their application as a hardmagnetic material. Despite the fairly large number of works dealing with the study of magnetic properties of the compounds with light interstitial atoms, these compounds still remain insufficiently studied (in particular, due to the complexity of obtaining single-crystal samples), though being extremely challenging objects for physics and technology of magnetic phenomena.

In this regard, it is of interest to investigate the effect of Sm<sub>2</sub>Fe<sub>17</sub> alloying particularly with titanium and molybdenum on the Curie temperature (T<sub>C</sub>) and magnetic properties.

#### 2. MATERIALS AND METHODS

To obtain the Sm<sub>2</sub>Fe<sub>17</sub> system alloy, powders of iron ( $d_{90} < 300 \mu m$ ) and samarium ( $d_{90} < 700 \mu m$ ) (Figure-1) as well as molybdenum and titanium with a purity of 99.9% were used as the initial components. Investigations were carried out on alloys of the following compositions:  $Sm_2Fe_{16}$ ,  $Sm_2Fe_{16}$ ,  $Sm_2Fe_{16}$ Ti,  $Sm_2Fe_{16}$ ,  $Sm_2Fe_{16}$ , SmSm<sub>2</sub>Fe<sub>16.5</sub>Ti<sub>0.25</sub>Mo<sub>0.25</sub>, and Sm<sub>2</sub>Fe<sub>16</sub>Ti<sub>0.5</sub>Mo<sub>0.5</sub>. Mechanical alloying was performed using Pulverisette-4 planetary mill with different rotation speeds according to the technique described in [20]. The duration of the mechanical alloying process was 20 and 40 hours. To prevent oxidation of the initial powder materials, preparation of powders for mechanical alloying and loading into mill bowls was carried out in a sealed glove box in a nitrogen atmosphere. The obtained powders were annealed in vacuum at a temperature of 700°C for 30 minutes.



**Figure-1.** The morphology of initial powders: a) iron, b) samarium.

Phase composition was studied by X-ray diffraction using Brucker D8 Advance diffractometer with © 2006-2016 Asian Research Publishing Network (ARPN). All rights reserved

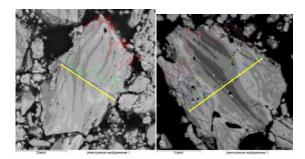


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 $CuK_{\alpha}$  radiation ( $\lambda$ =1,5418Å). The investigation of the structure and distribution of elements in the volume of powder particle was performed using scanning electron microscope Mira 3 Tescan with the Oxford INCA Wave 500 add-on device. Curie temperature (T<sub>C</sub>) was determined by the technique described in [21]. Studies of magnetic properties were performed on a vibrating magnetometer at the external field induction of 2.2 T.

### 3. RESULTS AND DISCUSSIONS

When conducting research on the effect of the mechanical alloying duration on the process of alloying elements dissolution, it was revealed that after 20 hours of mechanical alloying particles of the resulting powder constituted of a layered composite with a non-uniform distribution of alloying elements over the volume (Figure-2). alloying elements dissolution, it was revealed that after 20 hours of mechanical alloying particles of the resulting powder constituted of a layered composite with a nonuniform distribution of alloying elements over the volume (Figure-2). With increasing time of mechanical alloying the distribution of alloying elements over the volume straightens out and after 40 hours of synthesis all the alloying elements are uniformly distributed in the volume of powder and match the chemical makeup of the initial composition (Figure-3).



**Figure-2.** The distribution of elements in the volume of the powder particles obtained by mechanical alloying for 20 hours: a)  $Sm_2Fe_{16}Ti_{0.5}$ , b)  $Sm_2Fe_{16.5}Ti_{0.5}Mo_{0.5}$ .

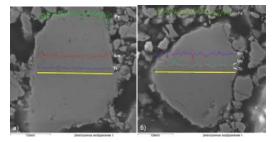


Figure-3. The distribution of elements in the volume of the powder particles obtained by mechanical alloving for 40 hours: a)  $Sm_2Fe_{16}Ti_{0.5}$ , b)  $Sm_2Fe_{16.5}Ti_{0.5}Mo_{0.5}$ .

The results of X-ray phase analysis of the studied powders have shown that the main phase component in the

samples is the phase with the crystal lattice corresponding to  $Th_2Zn_{17}$  type with a small content of  $\alpha$ -Fe, samarium oxide, and SmFe<sub>3</sub>. The addition of alloying elements results in a broadening and simultaneous shift of the peaks, belonging to Th<sub>2</sub>Zn<sub>17</sub>-type crystalline lattice (Figures 4 and 5) that is associated with the change in the crystalline lattice parameter when introducing alloying elements into the Th<sub>2</sub>Zn<sub>17</sub>-type lattice. The measurement results of lattice parameter are shown in Table-1. Presence of α-Fe and SmFe<sub>3</sub> suggests that the chemical reaction of Sm<sub>2</sub>Fe<sub>17</sub> formation was incomplete. The formation of samarium oxide, most likely, is caused by oxidation of samarium in the course of preparation of the initial powder composition or in the course of mechanical alloying due to the use of insufficiently pure gas.

During the investigation of the Curie temperature it was revealed that the additional alloying of the initial Sm<sub>2</sub>Fe<sub>17</sub> sample with titanium and molybdenum leads to the increase of the Curie temperature from 139°C (initial alloy) to 165-175°C (Table-2). Determination of magnetic characteristics was performed based on the parameters of the hysteresis loop. Table-2 shows the magnetic characteristics of the studied samples. According to the results obtained, the samples contain the fraction with anisotropy field exceeding 2.2 T; thus the loops turned out to be asymmetric (Figure-6). For the same reason, the values of the fore and reversal magnetization intensity are not the same. Doping of titanium, as well as molybdenum leads to a broadening of the hysteresis loop. The increase in the titanium content results in a slight increase in coercitive force at slight decrease in the residual magnetic induction (Table-2). Simultaneous doping of titanium and molybdenum has a greater impact on the increase of magnetic characteristics.

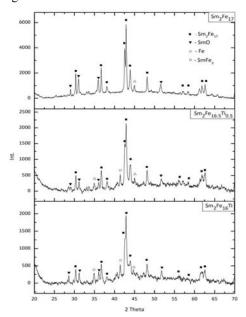
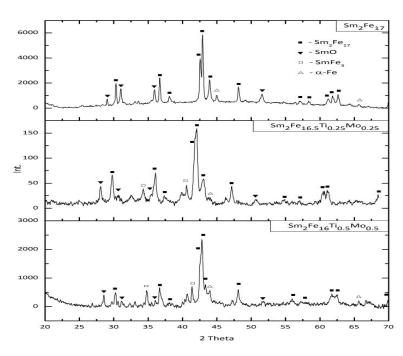


Figure-4. Changes in the phase composition of Sm<sub>2</sub>Fe<sub>17</sub> depending on the content of titanium.



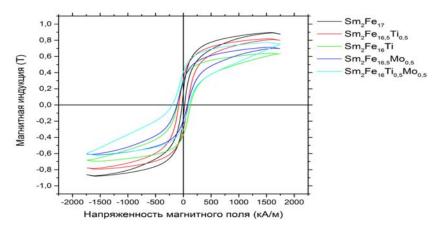
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**Figure-5.** Changes in the phase composition of Sm<sub>2</sub>Fe<sub>17</sub> depending on the content of titanium and molybdenum.

Table-1. Changes in the Sm<sub>2</sub>Fe<sub>17</sub> lattice parameter depending on alloying.

G '''	Lattice parameter					
Composition	a, Å	b, Å	c, Å			
Sm <sub>2</sub> Fe <sub>17</sub>	8,551	8,551	12,437			
Sm <sub>2</sub> Fe <sub>16.5</sub> Ti <sub>0.5</sub>	8,49371	8,49371	12,33321			
Sm <sub>2</sub> Fe <sub>16</sub> Ti	8,49371	8,49371	12,33321			
Sm <sub>2</sub> Fe <sub>16.5</sub> Mo <sub>0.5</sub>	8,48687	8,48687	12,38000			
Sm <sub>2</sub> Fe <sub>16.5</sub> Ti <sub>0.25</sub> Mo <sub>0.25</sub>	8,48683	8,48683	12,37468			
Sm <sub>2</sub> Fe <sub>16</sub> Ti <sub>0.5</sub> Mo <sub>0.5</sub>	8,50445	8,50445	12,33359			



**Figure-6.** The hysteresis loops for Sm-Fe-Ti, Sm-Fe-Mo, and Sm-Fe-Ti-Mo systems alloys obtained by mechanical alloying.

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**Table-2.** The magnetic characteristics of Sm-Fe-Ti, Sm-Fe-Mo, and Sm-Fe-Ti-Mo systems alloys obtained by mechanical alloying.

N	Alloy	Tc,°C	B <sub>r</sub> , T		B <sub>s</sub> , T		H <sub>c</sub> , kA/m		(BH) <sub>max</sub> ,
			up	down	-2T	<b>2</b> T	left	right	kJ/m <sup>3</sup>
1	Sm <sub>2</sub> Fe <sub>17</sub>	139	0,165	0,151	0,895	0,880	24,5	22,5	0,99
2	Sm <sub>2</sub> Fe <sub>16.5</sub> Ti <sub>0.5</sub>	166	0,330	0,307	0,817	0,793	89,8	78,4	7,67
3	Sm <sub>2</sub> Fe <sub>16</sub> Ti	175	0,310	0,370	0,639	0,695	109,3	123,4	10,00
4	$Sm_{2}Fe_{16.5}Mo_{0.5}$	165	0,289	0,187	0,618	0,615	115,3	74,1	8,41
5	$Sm_2Fe_{16}Ti_{0.5}Mo_{0.5}$	176	0,402	0,248	0,771	0,616	200,5	99,81	17,89

### 3. CONCLUSIONS

During research aimed at studying the effect of  $Sm_2Fe_{17}$  alloying, particularly, with titanium and molybdenum on the Curie temperature ( $T_C$ ) and magnetic properties. The alloying of  $Sm_2Fe_{17}$  with titanium and molybdenum leads to increase of the lattice parameter and the volume of the unit cell without changing the lattice symmetry. When measuring the hysteretic properties of  $Sm_2Fe_{17}$  based alloys obtained by mechanical alloying, it was revealed that the introduction of titanium, as well as molybdenum, leads to a broadening of the hysteresis loop. The simultaneous introduction of both titanium and molybdenum has a greater impact on the increase of magnetic characteristics.

### ACKNOWLEDGEMENTS

The work is carried out within the framework of the Federal target program "Research and development on priority directions of scientific-technological complex of Russia in 2014-2020" (the unique identifier of the project is RFMEFI57814X0087).

### REFERENCES

- [1] Tereshina I. S. 2003. Vliyanie legkikh atomov vnedreniya (vodoroda i azota) na magnitnuyu anizotropiyu i spin-pereorientatsionnye fazovye perekhody v intermetallicheskikh soedineniyakh 4f- i 3d-perekhodnykh metallov. Doktorskaya dissertatsiya. MGU, Moskva.
- [2] Zhang Z.D., Liu W., Liu J.P., Sellmyer D.J. 2000. Metastable phases in rare-earth permanent-magnet materials. Journal of Physics D: Applied Physics, 33: 217-246. PII: S0022-3727(00)09104-X.
- [3] Shen N.X., et al. 1999. Structural and magnetic properties of ammonia-nitrided Y<sub>2</sub>Fe<sub>17</sub>. Journal of Physics: Condensed Matter, 11: 833-845. PII: S0953-8984(99)96581-5.

- [4] Sugimoto S. 2011. Current status and recent topics of rare-earth permanent magnets. Journal of Physics D: Applied Physics. 44: 1-11. doi:10.1088/0022-3727/44/6/064001.
- [5] Ito M., Majima K., Shimuta T., Katsuyama S., Nagai H. 2002. Magnetic properties of Sm<sub>2</sub>(Fe<sub>0.95</sub>M<sub>0.05</sub>)<sub>17</sub>N<sub>x</sub> (M=Cr and Mn) anisotropic coarse powders with high coercivity. Journal of Applied Physics. 92(5): 2641-2645. DOI: 10.1063/1.1498880.
- [6] Bahr R., Hesse Jr.R., Boareto J.C., Wendhausen P.A. 2008. Sintering Sm<sub>2</sub>Fe<sub>17</sub> Prior to Nitrogenation. Materials Science Forum: 591-593, 75-79. DOI: 10.4028/www.scientific.net/MSF.591-593.75.
- [7] Nehdi I., et al. 2003. X-ray and Mössbauer studies of Sm<sub>2</sub>Fe<sub>17-x</sub>Cr<sub>x</sub> materials synthesized by mechanical alloying followed by an appropriate short annealing. Journal of Alloys and Compounds, 351: 24-30. PII: S0925-8388(02)01033-2.
- [8] Gutfleisch O., Clarke A.C., Neiva A.C., Sinan S.A., Harris I.R. 1996. Studies of HDDR processes in Sm<sub>2</sub>Fe<sub>17</sub>, Sm<sub>10.2</sub>Fe<sub>85.8</sub>Nb<sub>4</sub> and Sm<sub>9.5</sub>Fe<sub>80.5</sub>Nb<sub>10</sub> alloys. Journal of Alloys and Compounds, 233: 216-224. DOI:10.1016/0925-8388(95)02041-1.
- [9] Khazzan S., Bessais L., Van Tendelo G., Mliki N. 2014. Correlation between the nanocrystalline Sm(Fe,Mo)<sub>12</sub> and its out of equilibrium phase Sm(Fe,Mo)<sub>10</sub>. Journal of Magnetism and Magnetic Materials, 363: 125-132. http://dx.doi.org/10.1016/j.jmmm.2014.03.030.
- [10] Lee C.-H., Kwon Y.-S. 2002. Effect of mechanical alloying on the formation of Sm<sub>2</sub>Fe<sub>17</sub>N<sub>x</sub> compound. Metals and Materials International. 8(2): 151-154. DOI: 10.1007/BF03027011.

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- [11] Deng G., Jing Q., Wang X., He G., Ye X. 2010. Synthesis mechanism of Sm<sub>2</sub>Fe<sub>17</sub> alloy produced in reduction-diffusion process. Journal of rare earths, 28: 420-424. DOI: 10.1016/S1002-0721(10)60357-2
- [12] Gong W., Hadjipanayis G.C., Krause R.F. 1994. Mechanically alloyed nanocomposite agnets. Journal a Applied Physics. 75: 6649-6651. http://dx.doi.org/10.1063/1.356883.
- [13] Trosic J.T., Grujic A.S., et al. 2011. Magnetic behaviour of polymer bonded Nd-Fe-B composite materials. 15th International Research/Expert Conference. Trends in the Development of Machinery and Associated Technology. pp. 773-776. Prague, Czech Republic.
- [14] Lee J.-G., Kang S.-W., Si P.-Z., Choi A.-J. 2011. The influence of mechanical milling on the structure and magnetic properties of Sm-Fe-N powder produced by the reduction-diffusion process. Journal of Magnetics. 16(2): 104-107. DOI: 10.4283/JMAG.2011.16.2.104.
- [15] Saito T., Miyoshi H., Nishio-Hamane D. 2012. Magnetic properties of Sm–Fe–Ti nanocomposite magnets with a ThMn<sub>12</sub> structure. Journal of Alloys and Compounds. 519: 144-148. DOI:10.1016/j.jallcom.2011.12.156.
- [16] Ivanova G.V., Makarova G.M., Markin P.E., Popov A.G., Gorbunov D.I. 2011. Phase composition and magnetic properties of phases in  $Sm_2(Fe_{1-x-y}Mn_xSi_y)_{17}$  Alloys (with  $0 \le x \le 0.1$  and  $0 \le y \le 0.3$ ). Physics of Metals and Metallography, 112 (4): 343–349. DOI: 10.1134/S0031918X11040223.
- [17] Ivanova G.V., Makarova G.M., Popov A.G., Belozerv E.V. 2009. Structure and magnetic properties of ironrich Sm-Fe-(V, W)-Ga alloys. The Physics of Metals and Metallography. 108(4): 341-346. DOI: 10.1134/S0031918X09100032.
- [18] Hiromasa Yabe, Toshiro Kuji. 2006. Crystal structure and its magnetization of rare earth–iron alloys by mechanical alloying. Journal of Alloys and Compounds: 408-412, 313–318. doi:10.1016/j.jallcom.2005.04.056.
- [19] Yamamoto H., Osanai K. 2008. Magnetic properties of TbCu<sub>7</sub>-type Sm-Fe-Co-Mn-Ti-Y system nitrides. Journal of the Japan Society of Powder and Powder Metallurgy. 55 (7): 522–528. http://doi.org/10.2497/jjspm.55.522.

- [20] Popovich A.A., Razumov N.G., Verevkin A.S. 2015. Mechanical Alloying of Hard Magnetic Materials with Samarium. Applied Mechanics and Materials: 698, 339-344. doi:10.4028/www.scientific.net/AMM.698.339.
- [21] Determination of Curie point Temperature by TGA.
  Retrieved September 30, 2014.
  http://www.tainstruments.com/library\_download.aspx
  ?file=TS37.pdf.