Study of homogenization effect on the phase composition of $Sm_2(Fe, Co)_{17}$ alloys

UDC 621.318.12

V. P. Tarasov, *Head of Department of Non-Ferrous Metals and Gold, Professor*[1], *e-mail: vptar@misis.ru*
A. S. Ignatov, *Assistant, Department of Non-Ferrous Metals and Gold*[1]

[1] *National University of Science and Technology “MISiS”, Moscow, Russia.*

The aim of this work is elaboration of compositions of nanostructured hard magnetic materials (HMM) on basis of intermetallic compounds of the rare-earth metals (REM) with the iron-group transition metals, which posses the remanent magnetization (Br) no less than 1.20 T and coercive force (*Hci*) no more than 720 kA/m.

The emphasis of this paper has been on research of correlation between parameters of manufacturing process and actual structure of the high-coercive magnetic materials as well as on optimization of the phase composition of high-coercive nanostructured (HMM) based on nitrogen-containing alloys of REM. One of the basic means of increasing $Sm_2Fe_{17}N_3$ HMM magnet characteristics is lessening the $SmFe_3$ and $SmFe_2$ submagnetic phases content in magnetic medium, but chiefly the α-Fe presence, which leads to a drastic decrease of coercive force. And at the same time, the sole way to influence the process of phases distribution and formation in an ingot of magnetic material is optimization of the Sm_2Fe_{17} primary alloys melting practice. Optimal phase composition of $Sm_2(Fe, Co)_{17}N_3$ high-coercive HMM contains no less than 92% of the $Sm_2(Fe, Co)_{17}N_3$ basic magnet phase and no more than 0.5% of α-Fe phase.

It has been studied a phase co composition by value of the $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ alloy ingots, obtained by vacuum induction smelting and the following pouring into metal mould. As a result of the research it has been confirmed that phase composition of Sm_2Fe_{17} alloys, obtained by inductive smelting, has great heterogeneity which is mostly determined by the cooling rate. Effect of a high-temperature homogenization on the phase compositions of $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ alloys has been shown. The optimum temperature-time mode of homogenization has been determined.

Key words: hard magnetic materials, homogenization, permanent magnets, nitrogen-containing REM-Fe compounds, rare-earth metals, phase composition, nanostructured materials, magnetic composite materials.

DOI: *10.17580/nfm.2016.12.09*

Introduction

At present, elaboration of magnets, based on the REM compounds with transition metals, is the most actively developing line in the field of permanent magnets. As these materials provide the unique magnetic characteristics, which exceed those of ferrite and cast magnets, the rare-earth metal-based magnets are irreplaceable in aircraft and motor-car construction, robotics, computers etc. [1].

Among all iron compounds with the rare-earth elements, the highest iron content and, hence, the highest saturation magnetization have R_2Fe_{17} compounds, where R stands for rare-earth elements and nitrides on their base, for example $Sm_2Fe_{17}N_3$ [2]. In actual practice, however, nitriding of Sm_2Fe_{17} alloys obtained by traditional smelting techniques, involves severe technological difficulties, caused by high content of the other phases ($SmFe_3$, $SmFe_2$, α-Fe), nitriding of which happens very slowly and doesn’t results in formation of hard magnetic materials.

Currently, there is a series of investigations concerning formation of phases in the Sm_2Fe_{17} alloys [3–11]. There has been established an influence of ingots cooling rates on their phase composition subject to equilibrium crystallization. However, the above mentioned works don’t touch upon a question of irregular phase distribution during Sm_2Fe_{17} alloys melting in real conditions of production using standard plant auxiliaries and typical manufacturing equipment.

The major task of this study has been an elaboration of the phase composition distribution in ingots of Sm_2Fe_{17} alloys, melted in UPPF–3M (УППФ–3М) industrial vacuum induction furnaces with the use of standard collapsible slit chill moulds with useful capacity of 35–40 kg.

Materials and research procedure

Alloys smelting has been carried out in a UPPF-3M vacuum-induction furnace. Iron, cobalt and part of samarium (10% of the total amount) have been put to the bottom of boron nitride crucible. The rest of samarium has been charged into batcher.

During heating, furnace charge has been degassed for 30 min at a temperature of 600–700 °C. Upon completion of the furnace charge degassing process, the useful capacity of the smelting furnace has been pumped out to the residual pressure of 0.1–0.2 Pa. After that, the melting chamber has been filled with argon up to the pressure of 0.6×10^5 Pa and the furnace charge has been brought to melting. Time of the melt holding in the liquid state was 3–4 min. Melt crystallization has been carried out by the melt pouring to collapsible chill mould weighing about 90 kg.

Homogenization of ingots has been fulfilled in SNVE-1.3.1 (СНВЭ-1.3.1) vacuum furnaces at a temperature of 1170 °C and residual pressure no more than 0.01 Pa.

Quantitative phase analysis of samples has been carried out on a DRON-3 (ДРОН-3) diffractometer. For handling the *X*-ray photographs, DRON and *X*-RAY program packages have been used. Quantitative phase determination has been realized by the use of a PHAN% software. Relative error of the method doesn't exceed 5% and is calculated automatically by program means for each definite phase and is issued with the results of analysis.

Research results

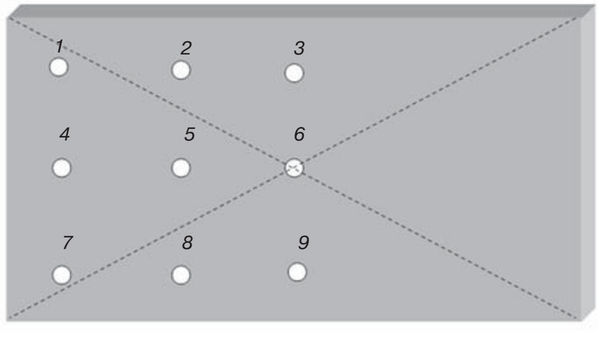
In the process of research there were smelted two ingots (weighing 35.6 and 35.9 kg) with identical charge composition. Results of chemical analysis of the ingots are summarized in Table 1.

The dimensions of the ingots were 50×350×550 mm. Samples have been taken out of the obtained ingots by drilling at a depth of 25 mm in accordance with the Figure.

Table 1

Chemical composition of $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ alloys

Ingot nomenclature	Content of primary elements, % (wt.)		
	Sm	Co	Fe
C1	24.7	8.1	Res.
C2	25.1	8.2	Res.



Scheme of samples taking for study of the $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ ingots phase composition

Table 2

Results of the quantitative phase analysis of $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ alloy samples

Sample number	Phase content, % (wt.)			
	Sm_2Fe_{17}	α-Fe	$SmFe_3$	$SmFe_2$
1	96.4±0.3	3.2±0.4	0.4±0.4	—
2	92.1±0.7	3.4±0.3	4.5±0.4	—
3	89.4±0.3	4.1±0.2	6.5±0.2	—
4	92.4±0.3	3.2±0.5	4.4±0.3	—
5	88.6±0.5	5.2±0.1	6.2±0.5	—
6	83.8±0.4	6.7±0.2	9.5±0.2	2.7±0.6
7	95.0±0.7	3.6±0.3	1.4±0.6	—
8	90.4±0.4	3.7±0.4	5.9±0.6	—
9	88.6±0.5	4.6±0.3	6.8±0.9	0.2±0.1

In the samples, there have been determined content of the primary phases, distinctive for Sm_2Fe_{17} alloys. In Table 2 are cited the data averaged through the results of examination of both ingots. Analysis of that table permits to draw a conclusion about sufficiently high heterogeneity of the primary phases distribution over the entire volume of the ingot.

As this take place, segments with the highest content of the main Sm_2Fe_{17} phase are situated along the edges of the ingot. Central part of the ingot contains the greatest amount of "ballast" phases, namely: α-Fe, $SmFe_3$ and $SmFe_2$, and is practically unusable as a material for manufacturing permanent magnets with high performance values.

In case of the repetition work, such segments of the ingots should be removed and sent to second melting. It should be also taken into account the fact that the samples have been obtained at a depth of 25 mm; that is in the central longitudinal plane of the ingot. Samples taken directly from the surface level of the ingot contains larger quantity of the major phase (by 2–3%), whereas α-Fe content drops by 25–30%.

In order to research a homogenization effect on the alloy phase composition, its ingots has been grinded to a size of 1.0–1.5 mm and blended. The necessity of the Sm_2Fe_{17} alloys homogenization is caused by the processes which take place on their crystallization out of the melt. The Sm_2Fe_{17} compound is forming at a temperature of 1177 °C [12] according to the peritectic reaction between gamma iron primary crystals and liquid phase. Since the melt cooling on the pouring happens quite rapidly, α-Fe crystals are always present in the ingot. If an unannealed material is used for the further manufacturing the $Sm_2Fe_{17}N_3$ magnetic material, then the present free iron will dissolve during nitration giving randomly oriented grains of the major magnetic phase, which leads to deterioration of magnetic parameters of magnets. Details of the homogenization influence on the Sm_2Fe_{17} alloy phase composition are represented in Table 3.

Table 3

Homogenization influence on the $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ alloy phase composition

Ingot nomenclature	Phase composition, %		Duration of homogenization, h
	Sm_2Fe_{17}	α-Fe	
Without homogenization	90.4	4.1	—
H1	91.7	3.9	16
H2	93.4	2.9	24
H3	95.6	1.8	32
H4	97.6	1.2	40

As can be seen from the Table 3, use of the alloys homogenization for 40 h allows to reduce the α-Fe phase content almost by a factor of 3.5. However, a prolonged ingot homogenization at 1170 °C for 40 h even in a deep vacuum (0.001 Pa) causes oxidation of material, and samarium oxide traces (up to 0.25% at 44 h of homogenization) appear in samples. Therefore, a homogenization durability of 36 h should be agreed as an optimum one.

Conclusions

1. Chemical and phase content of the $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ *industrial ingots, obtained by induction smelting in UPPF-3M furnaces and the following pouring into metal mould, is of a great heterogeneity over the volume.*

2. V*acuum homogenizing roasting of* Sm_2Fe_{17} *alloys for 40 h allows to equalize chemical and phase structure as well as to reduce content* of α-Fe *as an independent phase almost a 3.5 times.*

3. *Optimum* $Sm_2(Fe_{0.8}Co_{0.2})_{17}$ *alloy homogenization mode is determined by a temperature of 1170 °C and a 36-h holding.*

This article was written within the implementation of the Agreement on subsidies No. 14.578.21.0037 of July 22, 2014. (Unique identifier of the Agreement: RFMEFI57814X0037) between the National University of Science and Technology "MISiS" and the Ministry of Education and Science of the Russian Federation within the framework of the implementation of the Federal Target Program "Research and Development in the Priority Directions of the Scientific and Technological Complex of Russia Progress for 2014–2020", approved by the Decree of the Government of the Russian Federation of November 28, 2013, No. 1096.

References

1. Azzerboni B., Asti G., Pazeti L., Ghidini M. Magnetic Nanostructures in Modern Technology. Dordrecht : Springer, 2008. 345 p.

2. Sun H., Coey J. M. D., Otami Y., Hurley D. P. E. Magnetic properties of new series of rare-earth iron nitrides: $R_2Fe_{17}N_y$ ($y = 2, 6$). Journal of Physics: Condensed Matter. 1990. Vol. 2. pp. 6465–6470.

3. Nikitin S. A. *Magnitnye svoystva redkozemelnykh metallov i ikh splavov* (Magnetic properties of rare-earth metals and their alloys). Moscow : Moscow State University, 1989. 248 p.

4. Andreev A. V. *Magnitnye i magnitouprugie svoystva monokristallov soedineniy* R_2Fe_{17} (Magnetic and magneto-elastic properties of monocrystals of R_2Fe_{17} compounds). Fizika magnitnykh materialov : sbornik (Physics of magnetic materials : collection). Under the editorship of D. D. Mishin. Kalinin : State University, 1985. pp. 21–49.

5. Cou X. C. Magnetic anisotropy and magnetic phase transition in R_2Fe_{17} with R=Y, Ce, Pr, Nd, Sm, Gd, Tb, Ho, Er, Tm and Lu. Journal of Magnetism and Magnetic Materials. 1988. Vol. 177. pp. 1002–1007.

6. Coey J. M. D. Rare-earth iron permanent magnets. Oxford : Clarendon Press, 1996. 178 p.

7. Gabay A. M. Hadjipanayis G. C. Stability of $PrCo_{5+\mathrm{delta}}$ and $Pr_{1-x}Hf_xCo_{5+\mathrm{delta}}$ alloys with $TbCu_7$-type structure at 900–1150 degrees C and their magnetic properties. Journal of Alloys and Compounds. 2011. Vol. 509, No. 5. pp. 2562–2566.

8. Arinicheva O. A., Lileev A. S., Reissner M., Kubel F., Dormidontov A. G. Effect of Homogenization Temperature on the Structure and Properties of Sintered Magnets Based on Sm(Co, Fe, Cu, Zr)z. Metal Science and Heat Treatment. 2015. Vol. 56, No. 11–12. pp. 595–598.

9. Arinicheva O. A., Lileev A. S., Reissner M., Dormidontov A. G. Effect of heat treatment modes on the structure and magnetic properties of sintered permanent magnets based on Sm(Co, Fe, Cu, Zr)z. Metal Science and Heat Treatment. 2013. Vol. 55, No. $^{1}/_{2}$ pp. 63–67.

10. Chaubey G. S., Poudyal N., Liu Y. Z., Rong C. B., Liu J. P. Synthesis of Sm – Co and Sm – Co/Fe nanocrystals by reductive annealing of nanoparticles. Journal of Alloys and Compounds. 2011. Vol. 509, No. 5. pp. 2132–2136.

11. Zhang W. C., Zhao R., Fang Y. K., Zhou M. G., Zhu M. G., Li W. Effect of Sm-rich liquid phase on magnetic properties and microstructures of sintered 2:17-type Sm – Co magnet. Journal of Rare Earths. 2011. Vol. 29, No. 10. pp. 934–938.

12. Diagrammy sostoyaniya dvoynykh metallicheskikh sistem : spravochnik (Constitution diagrams of double metallic systems : reference book). In three volumes. Under the editorship of N. P. Lyakishev. Moscow : Mashinostroenie, 1997. 1024 p. NFM