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Structure and magnetic properties of $Sm_2Fe_{17}N_x$ alloys after severe plastic deformation by high pressure torsion

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ABSTRACT

In this study the structure and magnetic properties of $Sm_2Fe_{17}N_x$ alloys after high pressure torsion (HPT) was investigated. As a result of HPT, bulk samples were obtained as disk-shaped 0.5 mm high and 8 mm in diameter. The maximum coercivity H_c = 621.4 kA/m (7.7 kOe) was obtained on the samples after 3 turns (e = 5.01) with the remanence magnetization σ_r = 57.7 A·m^2/kg. It was shown that HPT leads to decomposition of the $Sm_2Fe_{17}N_x$ phase with the formation of α-Fe and the exchange-coupled state. The influence of temperature on the deformation processes of $Sm_2Fe_{17}N_x$ alloys was established.

1. Introduction

From the moment of discovery [1,2] to the present, the Sm_2Fe_{17} nitrides are the most promising materials for producing permanent magnets with higher magnetic properties than the Nd-Fe-B alloys due to the higher Curie temperature 470 °C [3], high magnetocrystalline anisotropy constant, an anisotropy field of 14 T [3], better oxidation resistance, lower rare earth (RE) metals content and lower cost of Sm compared to other RE metals. However, this system has few disadvantages, among which is the complexity of the nitriding process through the gas phase, the low decomposition temperature of $Sm_2Fe_{17}N_x$ up to 600 °C [3]. That does not allow obtaining magnets by sintering. High pressure torsion deformation is one of the methods that allows to obtain bulk nanostructured materials due to the formation of a high defect density in the crystalline structure. The formation of a crystalline texture is observed often when using this method [4 –6] and this is extremely important for obtaining high magnetic properties. In this regard, the study of the structure and magnetic properties of $Sm_2Fe_{17}N_x$ alloys after HPT is an urgent task.

2. Material and methods

Alloys of the Sm-Fe system were obtained from pure components (99.9% purity) by the vacuum induction melting technique. Then they were exposed to homogenization annealing in vacuum at 1100 °C for 40 h. Nitriding of the alloys was carried out after a double hydrogenation-dehydrogenation process at 400 °C for 40 h at a nitrogen pressure of 15 atm. The nitrogen content in the $Sm_2Fe_{17}N_x$ compound was determined by measuring the nitrogen pressure during nitriding and also by changing the lattice spacings of the $Sm_2Fe_{17}N_x$ phase. HPT was carried out on $Sm_2Fe_{17}N_x$ alloy powders in a Bridgman chamber at room temperature with pressure of 6 GPa and a number of turns n = 3 and 5 with a speed of 1 rpm. The true logarithmic strain *e* upon severe plastic deformation at the edge of sample was determined according to [7]:

$$e = ln\left(1 + \left(\frac{\varphi \cdot r}{h}\right)^2\right)^{0.5} + ln\left(\frac{h_0}{h}\right) \quad (1)$$

where *r* and *h* are the radius and the height, respectively, of the disk sample, and φ is the angle of anvil rotation. The *e* value in accordance with (1) was varied approximately between 5.01 and 5.54. Samples after HPT were disk-shaped with a diameter of 8 mm and a thickness of 0.5 mm. X-ray diffraction (XRD) analysis was performed on a Rigaku Ultima IV diffractometer using Co-K_α radiation. Spectrum analysis was performed using PDXL software (Rigaku). The microstructure of the alloys was studied using a TESCAN VEGA

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3 SBH electron microscope. The magnetic properties were measured on a physical properties measurement system (PPMS) (by Quantum Design) facility using vibrating sample magnetometer (VSM) device in the field 7.2 MA/m (90 kOe). Magnetic properties were measured on bulk samples and the data were corrected for demagnetization factor.

3. Results and discussion

According to XRD (Fig. 1) analysis, the samples after melting and homogenization are is a single-phase state and contain the Sm_2Fe_{17} (R-3m) phase. Nitriding leads to an increase of the lattice spacing, which indicates the dissolution of nitrogen in the Sm_2Fe_{17} phase. According to the change in nitrogen pressure during nitriding and an increase in the cell volume, it is revealed that the nitrogen content: x = 2.85 nitrogen atoms per formula unit. So the alloy can be described by the stoichiometric formula $Sm_2Fe_{17}N_{2.85}$. The XRD phase analysis of the $Sm_2Fe_{17}N_{2.85}$ samples showes that HPT at room temperature is accompanied by the decomposition of the $Sm_2Fe_{17}N_{2.85}$ phase into α-Fe (Im3m) and SmN (Fm3m). Moreover, the amount of decomposition products increases with increasing degree of deformation and reaches maximum values about of 35% α-Fe and 5% SmN at n = 5 turns. According to the measurement of the lattice spacings of the $Sm_2Fe_{17}N_x$ phase, HPT leads to a decrease in the cell volume from 843 $Å^3$ to 810 $Å^3$, which indicates a decrease in the nitrogen concentration from 2.85 to about 1.1 nitrogen atoms per cell unit. An analysis of the relative intensity of the XRD pattern peaks showed that the diffraction patterns of the samples after deformation does not reveal any features of the crystalline texture formation. According to the analysis of the diffraction peaks broadening, the process of HPT is accompanied

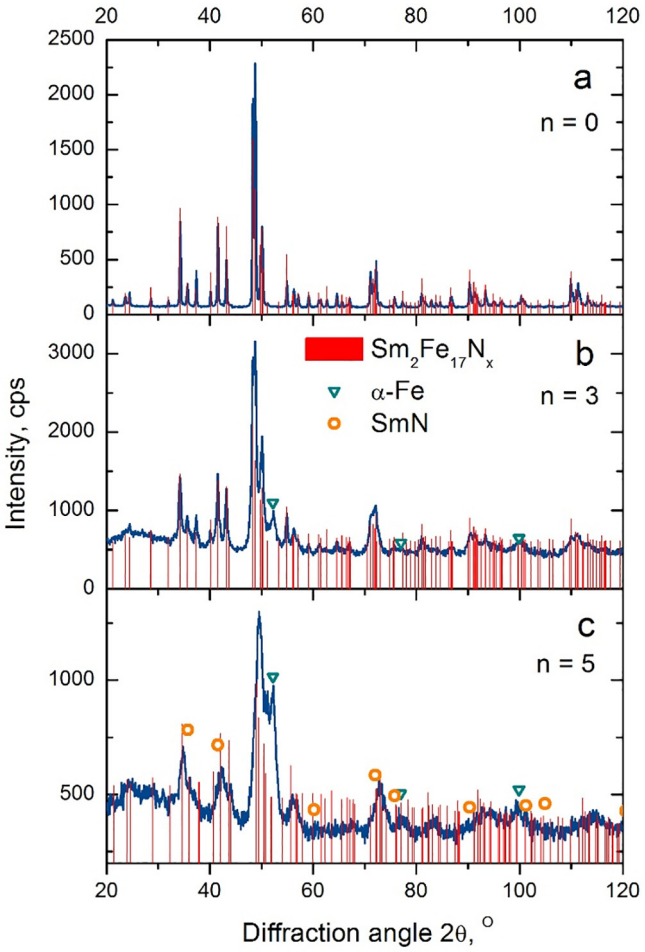

Fig. 1. XRD patterns of $Sm_2Fe_{17}N_x$ alloys after HPT with number of rotations n = 0 (a), 3 (b), 5 (c).

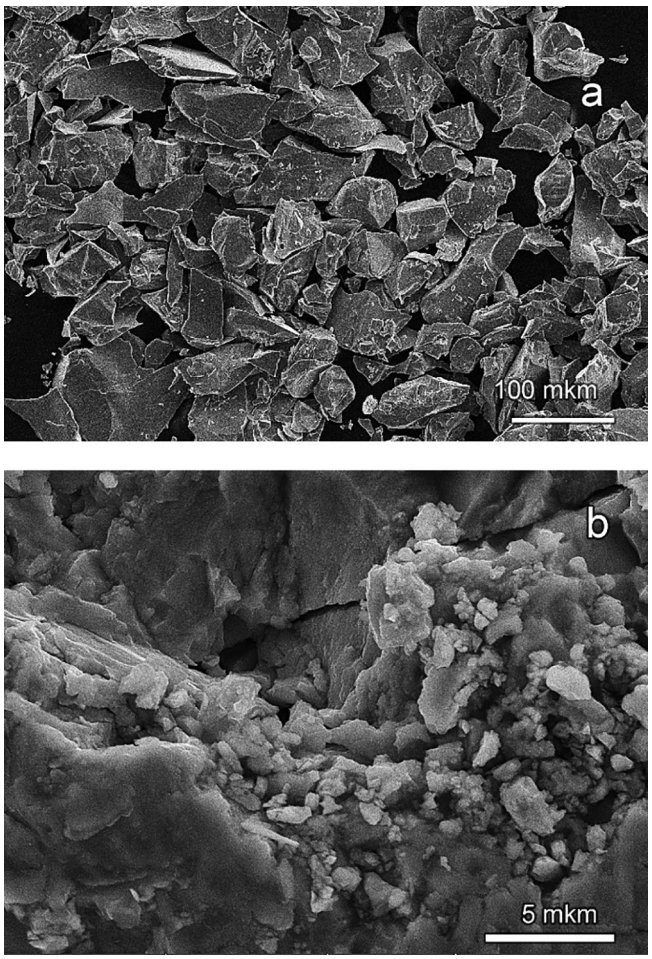

Fig. 2. Microstructure of $Sm_2Fe_{17}N_x$ alloys before (a) and after (b) HPT with n = 5 rotations.

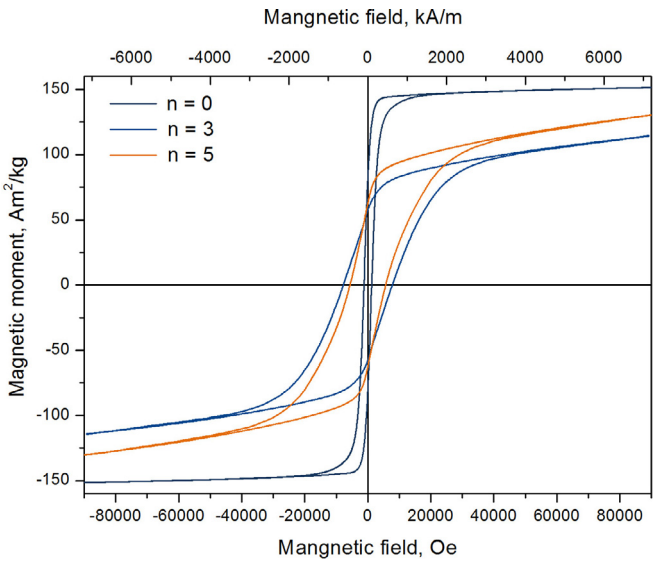

Fig. 3. Magnetic hysteresis loops of $Sm_2Fe_{17}N_x$ alloys after HPT.

Table 1
XRD phase analysis results and magnetic properties of $Sm_2Fe_{17}N_x$ alloys after HPT.

Turns, n	Phase composition			Lattice spacings of $Sm_2Fe_{17}N_x$, nm	Magnetic properties		
	$Sm_2Fe_{17}N_x$	α-Fe	SmN		H_c, kA/m (kOe)	σ_r, A·m²/kg	σ_s, A·m²/kg
0	100	–	–	a = 0.8757 c = 1.2694	98.5 ± 0.5 (1.232)	81.5 ± 0.2	151.4 ± 0.2
3	95 ± 2	5 ± 1	–	a = 0.8743 c = 1.2674	621.4 ± 0.5 (7.768)	57.7 ± 0.2	114.5 ± 0.2
5	60 ± 3	35 ± 3	5 ± 2	a = 0.8624 c = 1.258	451.1 ± 0.5 (5.639)	62.5 ± 0.2	130.2 ± 0.2

by a decrease in crystallite size down to 20 nm. The data of XRD analysis is confirmed by the results of scanning electron microscopy, which are presented in Fig. 2. The particle size after nitriding is several tens of micrometers, while after HPT with n = 5 turns the size of the structural elements decreases to a level of 100 nm.

Hysteresis loops of $Sm_2Fe_{17}N_x$ alloys after HPT are shown on Fig. 3, and magnetic properties are shown in Table 1. According to these results, the coercivity increases with an increase in the degree of deformation and reaches a maximum of H_c = 624.4 kA/m (7.768 kOe) at n = 3. A further increase in the degree of deformation at n = 5 leads to a decrease in the coercivity down to H_c = 451.1 kA/m (5.639 kOe) and is associated with the decomposition of the main hard magnetic phase accompanied by an increase of the α-Fe and SmN content. All loops after HPT do not have kinks, which indicates on the presence of an exchange coupled state between the $Sm_2Fe_{17}N_x$ and α-Fe phases. The saturation magnetization of alloys after deformation increases with an increase of n, which is associated with a greater magnetization of α-Fe comparing to $Sm_2Fe_{17}N_x$. Remanence, like saturation magnetization, increases with an increase in the degree of deformation n and exceeds 0.55, which also confirms that the exchange coupling bias presents between the phases $Sm_2Fe_{17}N_x$ and α-Fe.

As already noted, the decomposition temperature of the $Sm_2Fe_{17}N_x$ phase is about 600 °C [3], however, in this work decomposition during HPT is observed at room temperature. In our previous work [8], it was found that only refinement of the structure parameters is observed after HPT of $Sm_2Fe_{17}N_x$ alloys at 77 K without changing in the phase composition. Thus, large external forces acting during the HPT process can cause phase transformations, namely, the decomposition of the $Sm_2Fe_{17}N_x$ phase and significant refinement of the structure parameters. Due to the accumulation of a high density of structural defects during HPT at room temperature, diffusion processes that correspond to a higher temperature on the phase diagram begin to occur. Summarizing the results of this work and [8], the effective temperature at which the material is under HPT can be estimated. If no decomposition is observed during deformation at 77 K, then an effective temperature increase does not exceed about 800 K; while deformation at room temperature corresponds to processes at temperature that is 600 K higher. Thus, an effective temperature increase is 700 ± 100 K. This result goes in a good agreement with the published data [9,10], especially in similar systems [11,12].

4. Conclusions

In this work bulk $Sm_2Fe_{17}N_x$ samples obtained by the high pressure torsion (HPT) technique have maximum magnetic properties H_c = 624.4 kA/m (7.768 kOe), σ_r = 57.7 A·m²/kg, σ_s = 114.5 A·m²/kg with a number of turns n = 3. During the HPT the $Sm_2Fe_{17}N_x$ phase decomposites into α-Fe and SmN phases, which leads to the formation of an exchange-coupled state. The phase transformations occurring during HPT are equivalent to heating the material up to an effective temperature of 1000 K.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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