Application of spark plasma sintering for fabricating Nd-Fe-B composite

A A Sivkov [1], A S Ivashutenko [1] and A A Lomakina[1]
[1]Russia, Tomsk, Lenin ave, 30, National Research Tomsk Polytechnic University

E-mail: svechkanevaaa@mail.ru

Abstract. Constant magnets are applied in such fields as electric equipment and electric generators with fixed rotor. Rare earth metal neodymium is well known as promising material. Production of magnets by sintering three elements (neodymium, iron and boron) is one the most promising methods. But there are difficulties in choosing the right temperature for sintering and further processing. Structure and properties of the product, consisted of rare earth metals, was analyzed. X-ray analysis of the resulting product and the finished constant magnet was performed. Vickers microhardness was obtained.

1. Introduction

The permanent magnets (PM) are widely used in various fields of science and technology, such as radio and electrical engineering, electronics, devices for recording information, microelectronics, etc. The advantages of the permanent magnets are as follows: long life, material savings, and reliability. The main aim of modern technology is to decrease sizes at the same time with the saving high operating characteristics. This requirement is also attributed to the PM. Now the most promising materials are magnets based on cobalt, neodymium and samarium. PM based on rare earth elements have a hexagonal structure, strong magnetic anisotropy and high Curie temperature.

In 1984 the compound of neodymium (Nd) and iron (Fe) was firstly mentioned [1]. The most successful materials in this group are based on cobalt and neodymium magnets. The main advantage of the neodymium-iron-boron magnets in comparison with the samarium-cobalt alloy is the price and the scale of production. Also such class of rare-earth compounds shows record values of residual magnetic induction B_r = 1.1-1.2 T and coercive force H_c = 950-1750 kA/m. Papers [2-4] show the possibility of obtaining a composite material Nd-Fe-B, having practical advantages, such as thermal stability, high corrosion resistance and fracture toughness. The mentioned above high values of magnetic characteristics can be explained by the fine structure of materials and they are produced with using high-intensity actions. Spark plasma sintering (SPS) can be attributed to these kinds of effects. One of the most important benefits of SPS-method is its high sintering speed, which can effectively restrain the grain growth and ensure the formation of a dense fine grain structure in a short time. This is the purpose of this paper.

2. Experimental

For sintering industrially produced nanopowders were used as precursors. Figure 1 shows scanning electron microscopy (SEM) images and X-ray diffraction patterns of powders: Nd (figure 1, a), Fe (figure 1, b) and B (figure 1, c). These precursors were used to obtain a mixture which was sintered by SPS method.

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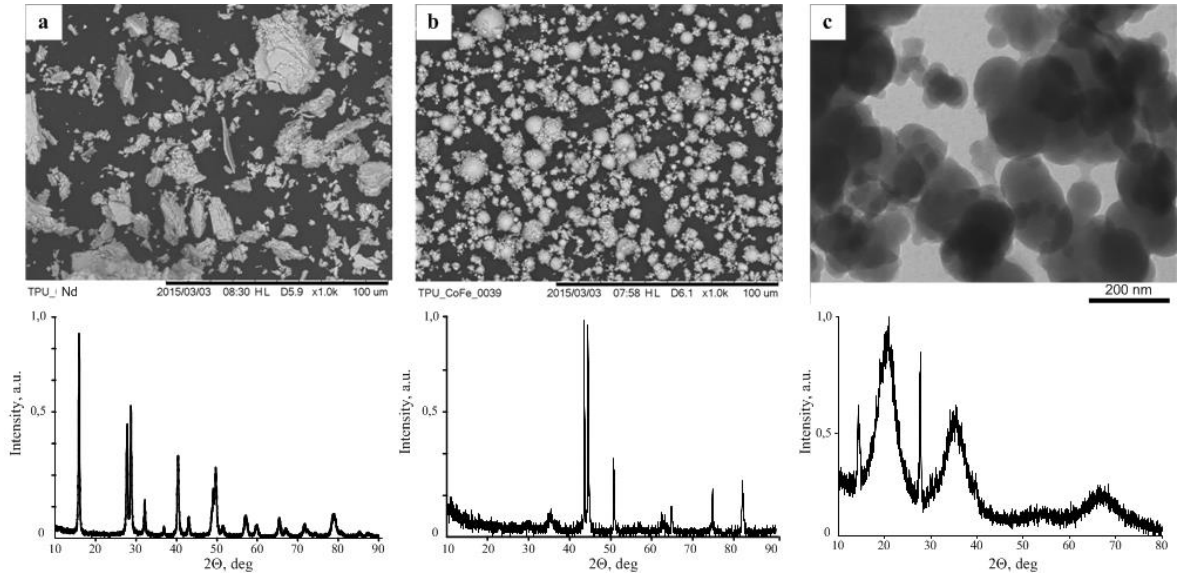

Figure 1. SEM-images and X-ray diffraction of industrial powders: neodymium (a), iron (b); TEM-image and X-ray pattern of boron (c).

The percentage of precursors in the mixture was as follows: Nd - 33 mass.%, Fe - 65 wt.%, B - 2 wt.%. The resulting mixture was mixed for 2 hours in a planetary mill SPEX Sample Prep. Mixer / Mill 8,000 M. After that the X-ray analysis for the powder mixture was carried out using diffractometer XRD Shimadzu 7000S. The XRD pattern is shown in Figure 2.

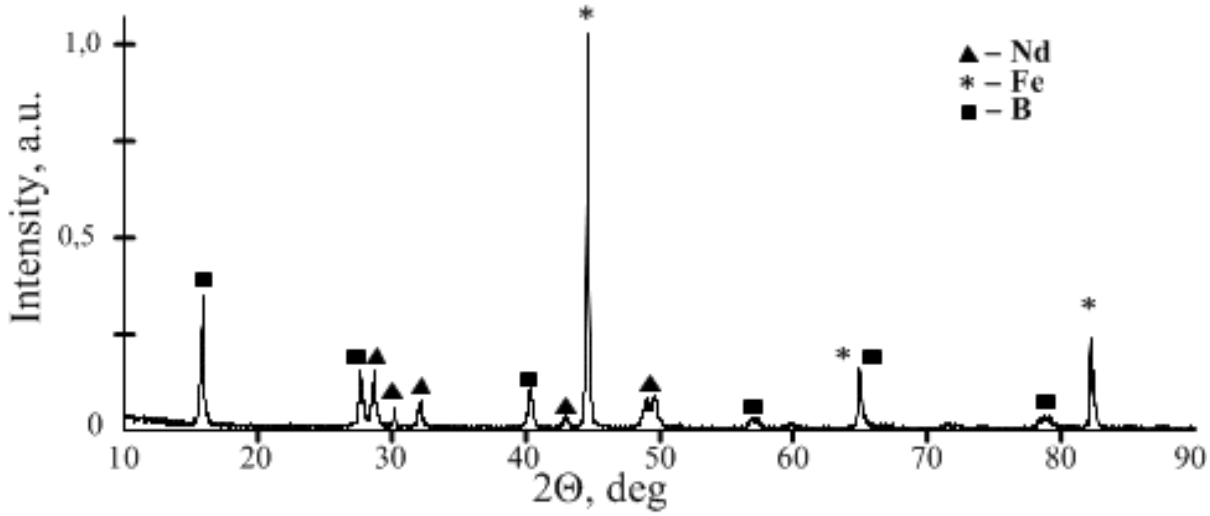

Figure 2. XRD pattern of mixture.

The finished mixture was sintered using the setup SPS10-4 (Advanced Technology, USA). The sintering was carried out in vacuum at 700 °C. The sintering mode was as follows: heating rate V_t= 100 °C/min, the pressing force 50 MPa, holding at the sintering temperature for 10 minutes, the current flow mode with a pulse repetition rate 25/5. The mixture material was molded in a graphite die with an inside diameter of 20 mm. The temperature monitoring was carried out using the K-type thermocouple, placed in the closed hole of the lower punch in 2.5 mm from the powder. The pressure was applied at the beginning of the sintering process and was removed after complete cooling of the sample.

Figure 3 shows the diagram of the geometric dimensions of the sample during sintering. The onset temperature of the sintering process (shrinkage) was 600 °C. In the time interval t_1-t_2 the sintering process went more intensively due to increasing temperature. In the case of isothermal hold at 700 °C the intensity of the shrinkage decreases.

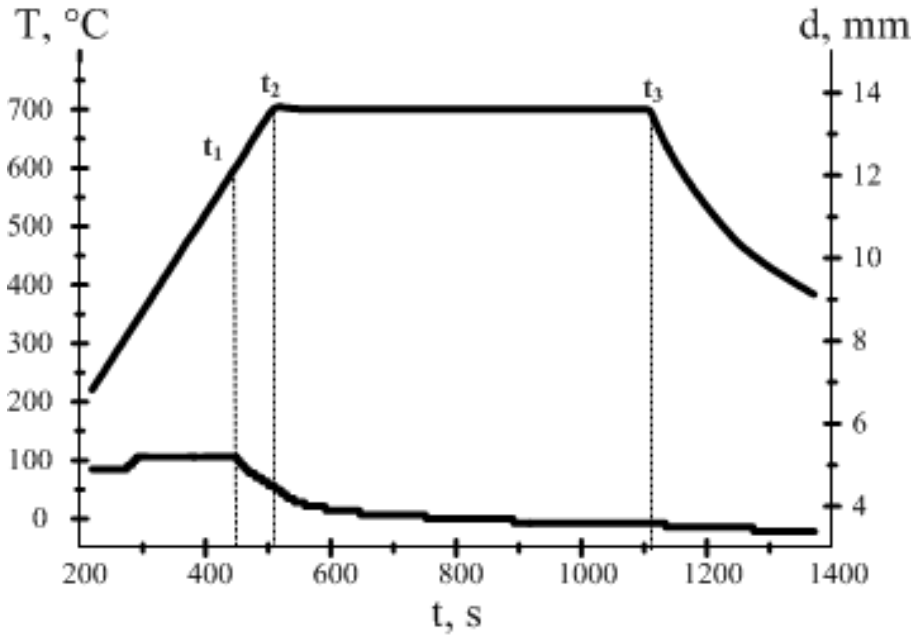

Figure 3. Diagram of the sintering process and the shrinkage of the sample during SPS-sintering.

After sintering, the sample was subjected to grinding. The hardness of the samples was studied using microhardness Isoscan OD HV2 Vickers. The microstructure of the polished surface and the chipping were examined using a scanning electron microscope HITACHI TM3000. The phase composition was determined using X-ray diffraction XRD Shimadzu 7000S (CuKα-radiation).

3. Results and discussion

The results of qualitative X-ray analysis were obtained using PDF2+. The figure 4 shows the phase analysis confirms the presence of such main PM phases as Nd, Fe and B.

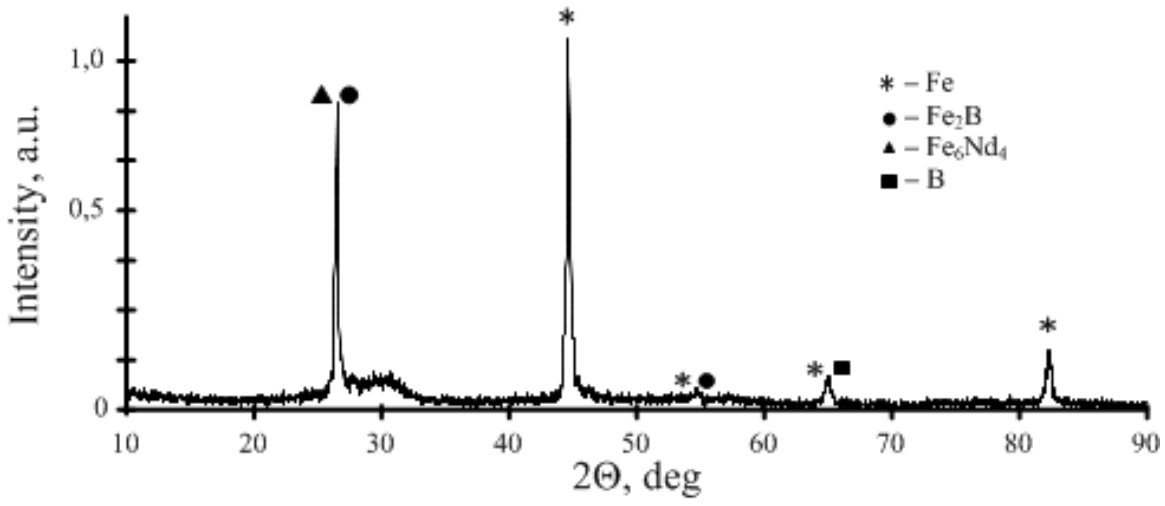

Figure 4. XRD pattern of Nd-Fe-B.

Data of the Vickers hardness and fracture toughness were obtained in the [5]. The hardness measurement was carried out using Vickers indenter according to the method of unrestored print. The average value of the microhardness was 142.06 HV. The maximum value of known hardness for permanent Nd-Fe-B magnets is 600HV. Also the average diagonal indentation equal to 0.1144 mm was measured.

Figure 5 shows the SEM-images of the surface of the sintered sample. The investigation of the sample surface showed the presence of sintered objects (Figure 5, b). In particular, there is an object not subjected to sintering and connections to other parts of the sample (see Figure 5, c and d).

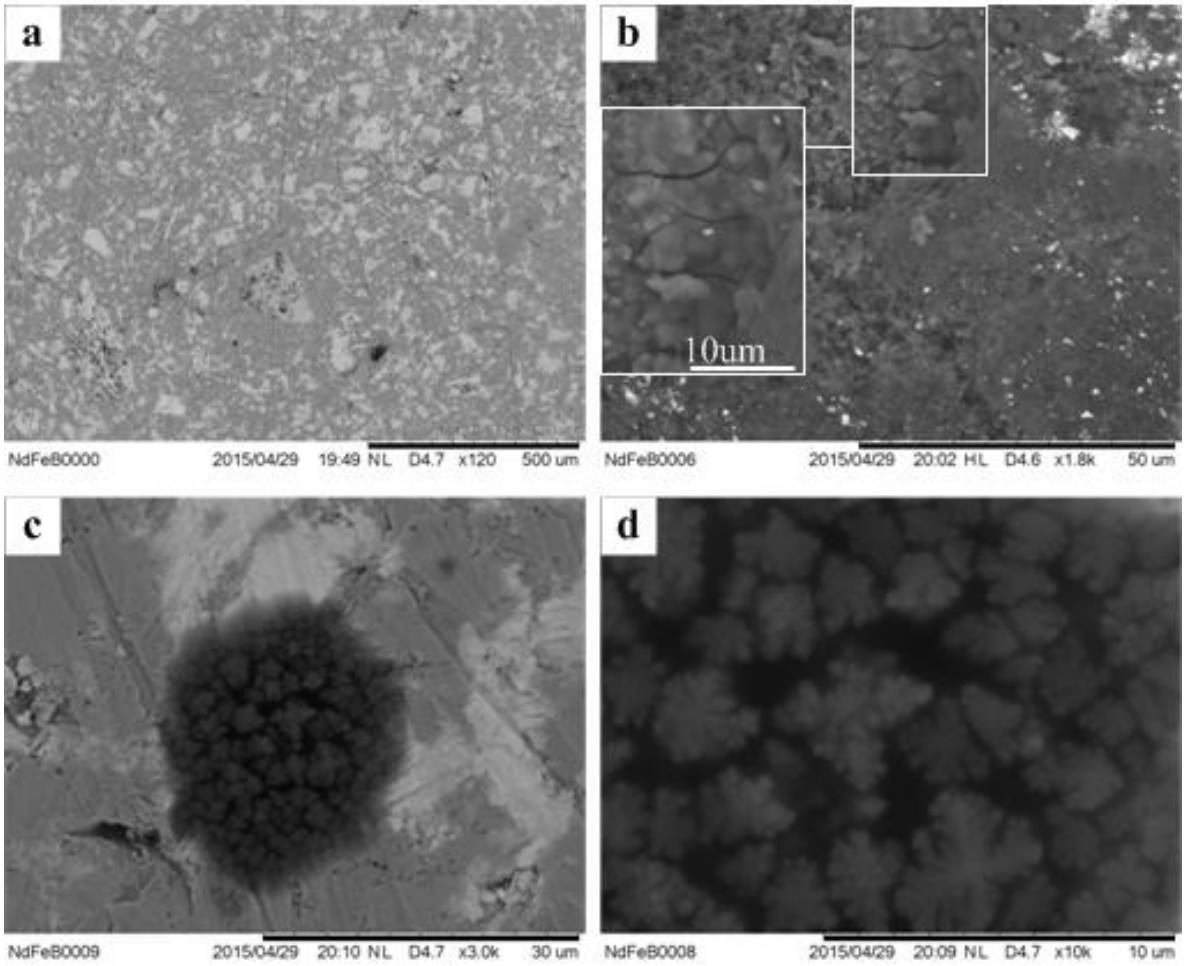

Figure 5. SEM-images of the sintered surface of the Nd-Fe-B sample.

In order to study the internal structure of the material the chipping was made. Figure 6 shows the SEM-images of chipping. An analysis of images allows us to conclude that the magnet material has a residual porosity. The density of the sample, which is equal to 6.32 g/cm^3, (theoretical density is 7.4 g/cm^3) was measured by hydrostatic method. Therefore, to obtain a dense structure it is necessary to increase the sintering temperature, and, possibly, the time of isothermal hold. The similar structure of the material is presented in [6,7].

To study the magnetic properties of the sample the magnetometer H-04 was used, taking the measurements in fields up to 30 kOe [8]. Figure 7,a shows the field dependence of the specific magnetization σ(H). The value of the specific magnetization is determined by the tangent line and is equal to 156 Gs·cm^3/g (dashed line in Fig. 9), which is in good agreement with published data (paper

[9] shows the value of specific magnetization of a permanent magnet $Nd_2Fe_{14}B$ with a value of more than 150 ° $Gs \cdot cm^3/g$).

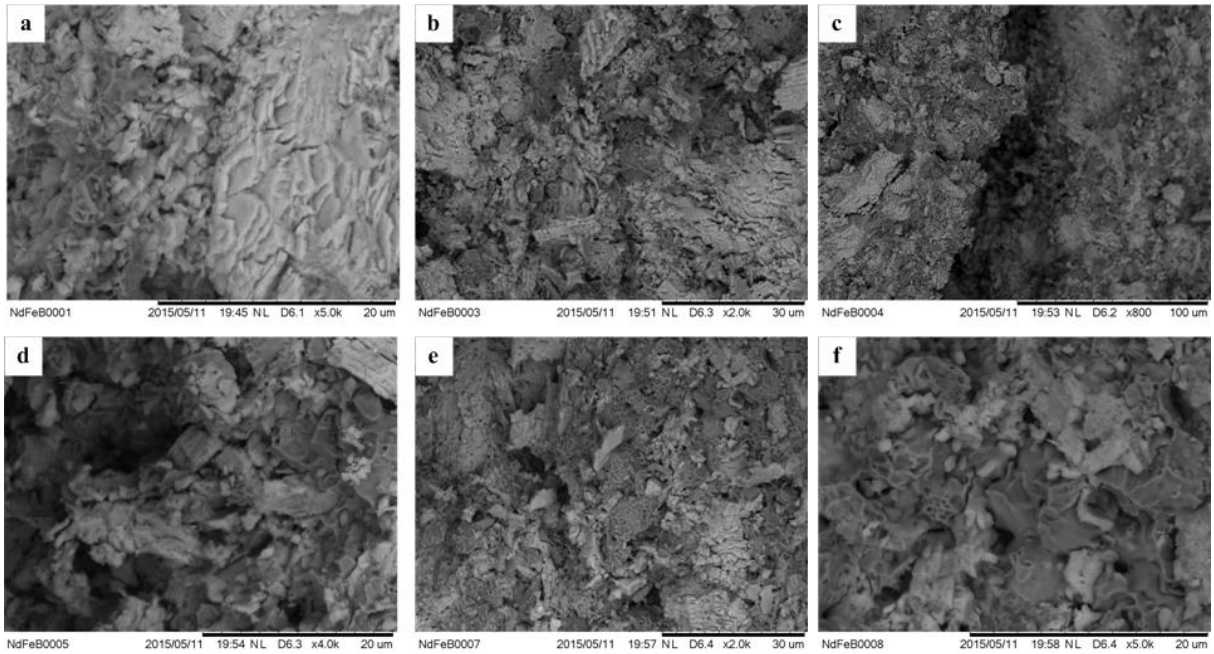

Figure 6. SEM-images of the chipping Nd-Fe-B.

The sample presented in our work has the effective anisotropy field at the level of 3,6 kOe (Figure 8). This value is less than that was previously described (6,4 kOe [2]). This difference can be explained by the residual porosity in our sample.

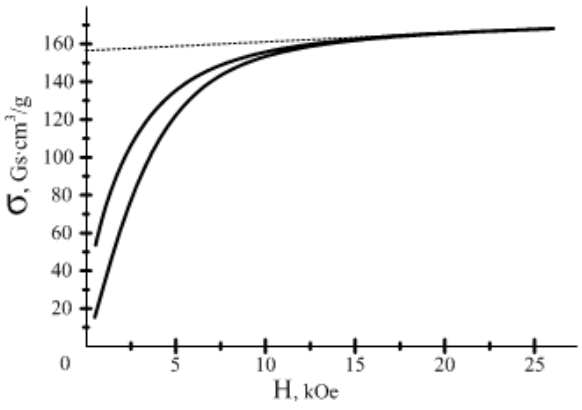

Figure 7. The curve of specific magnetization of SPS sample Nd-Fe-B

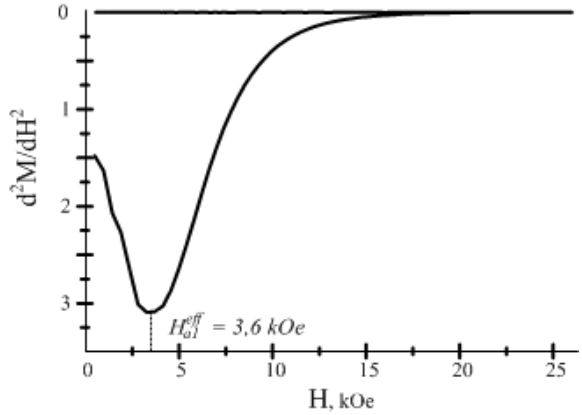

Figure 8. The curve of effective anisotropy field of SPS sample Nd-Fe-B

4. Conclusion

Thus the SPS-sintering method provides the synthesis of Nd-Fe-B permanent magnets in a forward and short process cycle (20 min.). This method allows synthesizing magnetic material, which combines high technology, the stability of the magnetic characteristics, relatively low cost. The experimental results provide further refinement of sintering parameters of magnetic material to improve its magnetic characteristics.

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