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Heat treatment process of new NdFeB magnet prepared by spark plasma sintering①

LI Tao(李 涛)[1], YUE Ming(岳 明)[2], ZHANG Jiu-xing(张久兴)[2],
WANG Gong-ping(王公平)[2], XIAO Yao-fu(肖耀福)[1], WANG Run(王 润)[1]
(1. School of Materials Science and Engineering,
University of Science and Technology Beijing, Beijing 100083, China;
2. Beijing University of Technology, Advanced Functional Materials Key laboratory,
Ministry of Education, Beijing 100022, China)

Abstract: In recent years, spark plasma sintering technique(SPS) has been a focus in the field of material preparation due to its advantages. SPS technique is first introduced for preparation of high quality NdFeB magnets. The effects of heat treatment process on the magnetic properties of SPS NdFeB magnet were investigated. Meanwhile, the effects of heat treatment process on the microstructure, tropism and dimensional precision of the SPS NdFeB magnets were also studied. The high quality NdFeB magnets with fine grains were prepared under proper heat treatment process. The results show that the magnetic properties of SPS NdFeB can be further improved through proper heat treatment process. Meanwhile, the experiment also demonstrates that it is feasible to prepare near-net-shape NdFeB magnets with fine grains and high magnetic property by spark plasma sintering.

Key words: spark plasma sintering; formability; shrinkage ratio; heat treatment

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1 INTRODUCTION

Traditionally sintered NdFeB magnet can meet the requirement of dimensional precision through post-machining, which usually results in 45% of material loss. Post-machining not only wastes the rare earth resources, but also increases the production cost. Moreover, it is difficult to prepare homogeneous workpieces with large dimension and complicated shape due to some uncontrollable factors in the conventional sintering process. On the other hand, although the bonded NdFeB magnet has better formability and dimensional precision, its application is greatly limited because of its unsatisfactory magnetic properties. Therefore, the Spark Plasma Sintering (SPS) technique[1] was introduced in this study for preparation of the NdFeB permanent magnet to find out the possibility of solving the problems mentioned above.

SPS is known as one of the novel sintering techniques. The SPS process utilizes the momentary local high temperature field generated by pulse energy, spark impact pressure and Joule heating throughout sintering to heat the specimen. Thus, it has many important advantages including lower sintering temperature and high sintering speed, which can effectively restrain the grains from growing and prepare fine-grained materials in relatively short time. Moreover, this kind of technique can also be used for the preparation of large-scale or complicate-shaped workpieces, e.g. magnet tiles and thin-wall magnet rings with high dimensional precision. Finally, the sintering process can be carried out under pressure, which is helpful for the sintered compact to obtain higher density. So far, SPS technique has been widely applied to the preparation of metallic materials, intermetallic compounds and ceramic materials[2-11].

The optimal magnetic properties haven't been obtained by NdFeB magnets prepared by SPS technique yet[12]. Their magnetic properties can be greatly improved through further heat treatment process. In this article, the effects of post heat treatment on the properties of SPS NdFeB magnet were investigated.

2 EXPERIMENTAL

Casting ingot with the composition of $Nd_{15.5}Dy_1Fe_{bal}Co_{3.0}B_{6.8}Al_{1.5}$ was prepared by vacuum in-

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Correspondence: YUE Ming, PhD; Tel: + 86-10-67392169; Fax: + 86-10-67392840; E-mail: yueming@bjpu.edu.cn

duction furnace. The ingot was then ground to powders with the average granularity of 5.0 μm by air-flow mill. Orientating and applying slight pressure to the magnetic powders in magnetic field after they have been enclosed in the graphite mould. The following sintering process was conducted on the SPS-3.20-MK-V type sintering machine, developed by Japanese Sumitomo Coal Mining Co, followed by cooling the specimen to the room temperature after the sintering process. The heat treatment was performed in a vacuum tube furnace. The same magnetic powder was also sintered using the conventional sintering process for better comparison.

The magnetic properties of the magnets were measured by CL-6 B-H tracer. The microstructure of the specimen was recorded using S-250MK Scanning Electron Microscope(SEM) and the composition analysis was conducted by Energy Dispersive X-ray Detector (EDX). The density of the specimen was measured by Archimedes method and the magnet size was measured by micrometer.

3 RESULTS AND DISCUSSION

3.1 Effect of heat treatment on magnetic properties of SPS NdFeB

3.1.1 Effect of heat treatment on magnetic properties of SPS NdFeB

The optimal magnetic properties haven't been obtained by SPS NdFeB yet. Their magnetic properties can be greatly improved through further heat treatment process, as shown in Table 1.

Table 1 Effect of heat treatment on magnetic properties of SPS NdFeB

Property	B_r/T	H_{ci}/(kA·m^{-1})	$(BH)_{max}$/(kJ·m^{-3})
SPS NdFeB	0.892	348	56
SPS NdFeB after heat treatment at 1 353 K for 2 h	1.105	838	193

3.1.2 Effect of temperature of first-order heat treatment on magnetic properties of SPS NdFeB

Temperature of heat treatment is an important condition for the thermodynamics and kinetics of carrying out phase change in the magnet. The first-order heat treatments on SPS NdFeB magnets were carried out for 2 h at different temperatures. The effect of the temperature of heat treatment on the magnetic properties is shown in Fig. 1, which indicates that the magnetic properties improve greatly with the rise of the temperature of heat treatment. However, the coercive force of the magnet drops evidently when the temperature is higher than 1 300 K. The effect of temperature of first-order heat treatment on the magnetic properties is related to the quantity, morphology and distribution of the Nd-rich phase. When the heat treatment is carried out at higher temperature, the Nd-rich phase at the grain boundary melts down into liquid in a short time. If the composition of the liquid Nd-rich phase can be optimized close to the Nd content in the eutectic reaction, the microstructure with higher coercive force can be obtained. From Fig. 1, it is clear that the comprehensive magnetic properties are the best when the temperature of first-order heat treatment is in the range from 1 250 K to 1 273 K. When the temperature of heat treatment is 1 323 K, the SPS magnet achieves the highest $(BH)_{max}$. However, too high heat treatment temperature (e. g. 1 353 K) is not conducive to the improvement of the magnetic properties of SPS NdFeB. Therefore, proper first-order heat treatment can effectively improve the magnetic properties of SPS NdFeB.

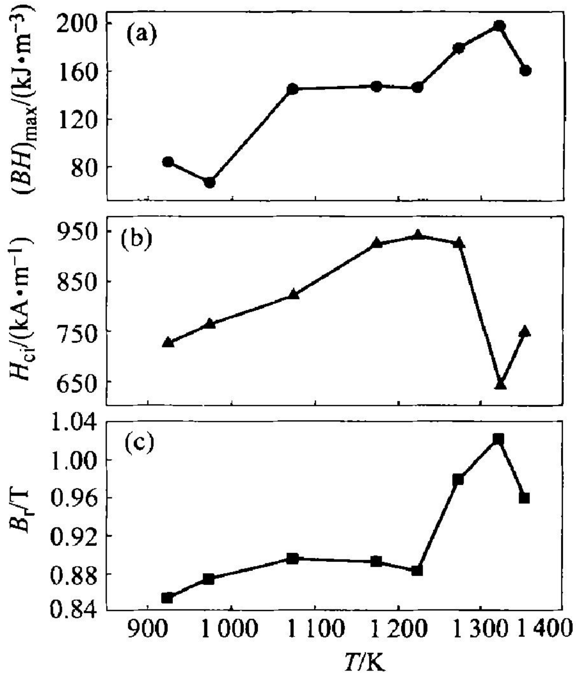

Fig. 1 Magnetic properties of SPS NdFeB as function of temperature of first-order heat treatment

3.1.3 Effect of temperature of second-order heat treatment on magnetic properties of SPS NdFeB

The second-order heat treatments on SPS NdFeB magnets were carried out for 1 h at different temperatures. The effect of the temperature of second-order heat treatment on the magnetic properties of the SPS NdFeB is shown in Fig. 2. In similar manner, the best temperature of second-order heat treatment is around 923 K.

3.2 Tropism study

The magnetic properties of sintered Nd-Fe-B permanent magnets mainly come from the tetragonal $Nd_2Fe_{14}B$ matrix phase. The tropism of the powder has great effects on the magnetic remanence and the

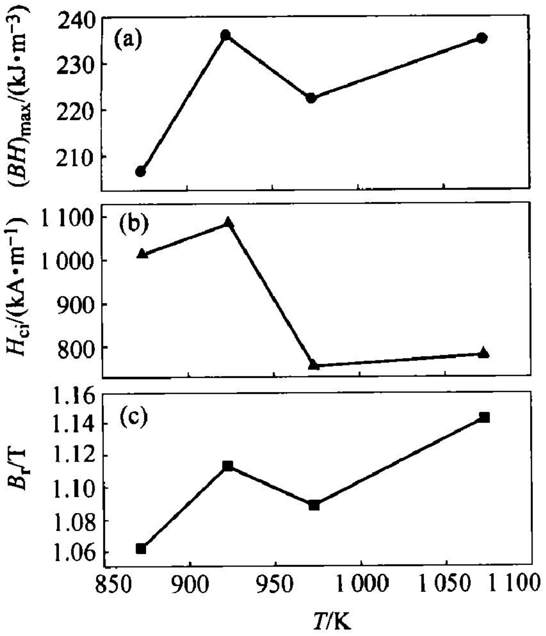

Fig. 2 Magnetic properties of SPS NdFeB as function of temperature of second-order heat treatment

energy product of the magnets. Therefore, powder tropism in the magnetic field is a key process in the preparation of high energy anisotropic sintered permanent magnets. The magnetic remanences in the parallel and vertical tropism directions of the anisotropic Nd-Fe-B permanent magnet are quite different. Therefore, the tropism *DOA* can be generally expressed in the following equation

$$DOA = \frac{B_{r\parallel} - B_{r\perp}}{B_{r\parallel}} \quad (1)$$

It is shown in Fig. 3 that the *DOA* of SPS NdFeB remains almost unchanged(only drops a little) after heat treatment. This demonstrates that the tropism can be retained in SPS and heat treatment processes, which is very important to the preparation of the anisotropic Nd-Fe-B permanent magnets.

3.3 Effect of heat treatment on microstructure of SPS NdFeB

The effect of heat treatment on the microstructure of the SPS NdFeB is shown in Fig. 4. It is clear from Fig. 4 that the heat treatment evidently betters the distribution of Nd-rich phase in SPS NdFeB and reduces the space and holes between the crystal grains, hence increases the density and the magnetic remanence of the magnet. The Nd-rich phase forms a net-like distribution along the grain boundary in Fig. 4(c) after heat treatment compared to the dispersed Nd-rich phase distribution in Fig. 4(a) before heat treatment. Meanwhile, the density of the SPS NdFeB increases from 7.37 g/cm^3 before heat treatment to 7.52 g/cm^3 after heat treatment. Moreover, SPS NdFeB has different morphology characteristics in

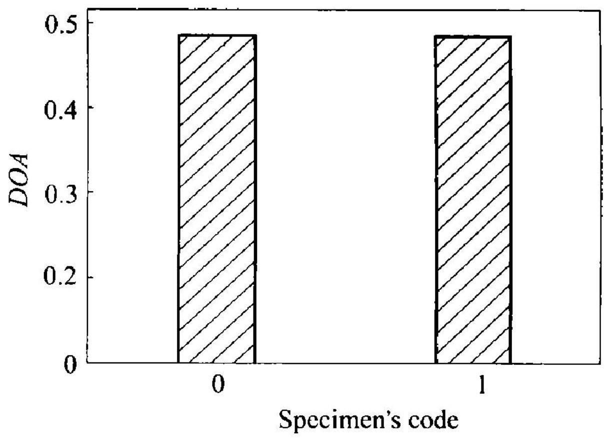

Fig. 3 Effect of heat treatment on *DOA* of SPS magnets

0 —SPS magnets; 1 —SPS magnets after first-order heat treatment at 1 273 K for 2 h

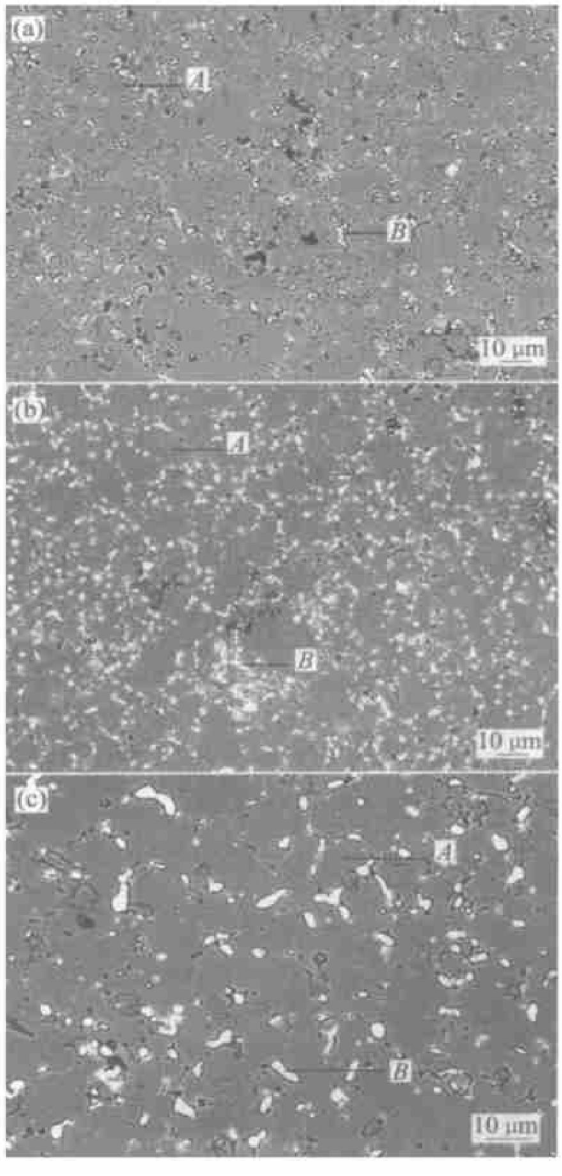

Fig. 4 Effect of heat treatment on microstructural characteristics of magnets (SEM Photos)

(a) —SPS; (b) —First-order heat treatment; (c) —Second-order heat treatment

A —$Nd_2Fe_{14}B$ phase; *B* —Nd-rich phase

Nd-rich phase distribution compared with that of traditionally sintered NdFeB. Therefore, heat treatment can greatly improves the magnetic properties, especially the coercive force of the magnets.

Another advantage is that the SPS technique can effectively restrain the grains from growing during sintering process and prepare fine grains materials, which is a main reason for the introduction of SPS technique into the preparation of Nd-Fe-B magnets in this study. Specifically speaking, the granularity thinning is an important way to improve the coercive force of the Nd-Fe-B magnets; meanwhile, the fine-grained microstructure can make more homogeneous composition distribution in the magnets, which is good for their anti-corrosive behavior. Fig. 5 shows the microstructure comparison of the SPS NdFeB and the traditionally sintered NdFeB after heat treatment. It can be seen from Fig. 5 that the granularity of SPS Nd-Fe-B magnet is about 5 − 10 μm after heat treatment and evenly distributed. The average grain size remains almost unchanged (as shown in Fig. 4). In comparison, the granularity for the traditionally sintered Nd-Fe-B is relatively larger by about 20 μm and not evenly distributed (Fig. 5). From the experiment results obtained so far, the fine grained SPS Nd-Fe-B with higher magnetic properties has been prepared after proper heat treatment. Meanwhile, it is notable that the Nd-rich phase distribution of SPS Nd-Fe-B is different from that of traditionally sintered Nd-Fe-B.

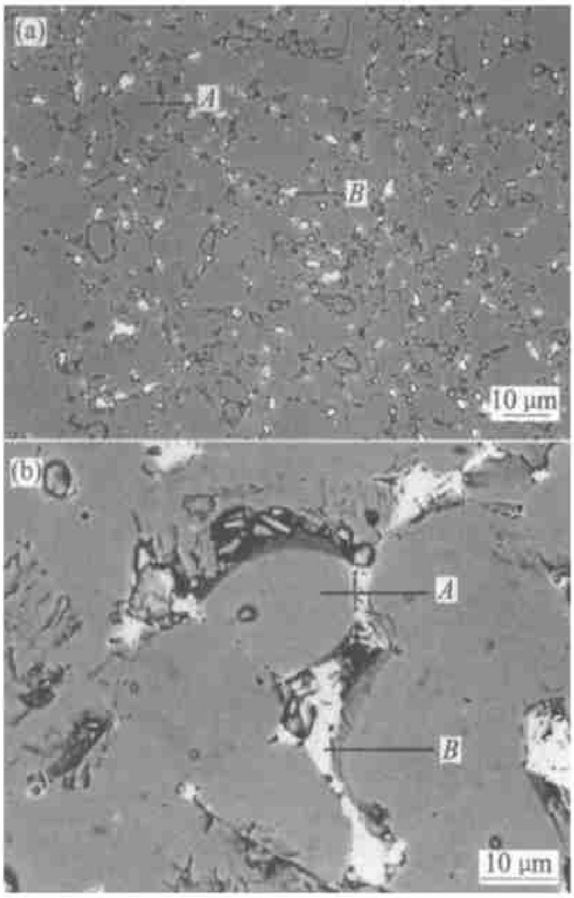

Fig. 5 Comparison of SPS NdFeB and traditionally sintered NdFeB (SEM photos)
(a) —SPS NdFeB after first-order heat treatment at 1 353 K for 2 h;
(b) —Traditionally sintered NdFeB
A —$Nd_2Fe_{14}B$ phase; *B* —Nd-rich phase

3.4 Effect of heat treatment process on dimensional precision of SPS NdFeB

Ideal dimensional precision is the key factor to the application of the near net shape technique in the preparation of the high performance NdFeB magnet. Therefore, the shrinkage ratios before and after the heat treatment are investigated in the study. The corresponding densities of the magnets are also listed in Table 2 for the convenience of reference.

Table 2 Change of shrinkage ratio and density of SPS NdFeB before and after heat treatment

Property	Shrinkage ratio/ %	Density / ($g\cdot cm^{-3}$)
SPS NdFeB	/	7.00
SPS NdFeB after heat treatment at 1 073 K	0.5	7.10
SPS NdFeB after heat treatment at 1 273 K	1.0	7.17
SPS NdFeB after heat treatment at 1 353 K	4.0	7.58

The dimensional deviation of SPS NdFeB in the same dimension is less than 20 μm, which is even better than that of the machined conventionally sintered NdFeB. Therefore, the SPS NdFeB magnet has excellent dimensional precision. It is shown in Table 2 that SPS NdFeB can have relatively small shrinkage ratio after heat treatment at proper temperature. Therefore, proper heat treatment can increase the density of the SPS NdFeB, greatly improve the magnetic properties of SPS NdFeB as well as retain better dimensional precision.

4 CONCLUSIONS

1) Proper heat treatment process can effectively improve the magnetic properties of SPS NdFeB. The microstructure of SPS NdFeB is different from that of the traditionally sintered one.

2) After proper heat treatment, the average granular size of SPS NdFeB only changes slightly. Therefore, the SPS technique is promising to be an effective approach for the preparation of NdFeB magnets with fine grains.

3) Proper heat treatment can increase the density of the SPS NdFeB, greatly improve the magnetic properties of SPS NdFeB as well as retain better dimensional precision. It is shown that the near net

shape SPS technique is feasible for the preparation of the high performance NdFeB magnet.

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